

SESSION 2

Chairman: Professor M. SALVATORES (France)

THE FRENCH PARTITIONING-TRANSMUTATION PROGRAMME,
ASSETS AND PROSPECTS

Michèle VIALA, Commissariat à l'Energie Atomique, Fuel Cycle Division
Massimo SALVATORES, Commissariat à l'Energie Atomique, Nuclear Reactor Division
Henri MOUNEY, Electricité de France, Equipment Division

ABSTRACT

Partitioning-transmutation studies are covered by the 1991 French law concerning radioactive waste management.

The programme is progressing with a dual approach:

- What can be done in partitioning-transmutation? At what cost? In what timescale?
- How can long-term gains and short-term disadvantages be qualified and quantified?

The first approach concerns technical know-how. The studies based on today's technologies are continuing (reactors, fuels and targets, separation of radionuclides by solvents).

The second approach involves an assessment activity, based firstly on studies of scenarios. Pertinent assessment criteria must be brought out.

INTRODUCTION

The French law of December 1991 on radioactive waste management requires this management to comply with the protection of nature, environment and health, taking the rights of future generations into consideration. It explicitly indicates that solutions must be found to enable the partitioning and transmutation of the long-lived radionuclides present in these wastes.

In 2006, the French Parliament will have to come to a decision as to the construction of an underground disposal and the continuation of other research paths.

The French Assessment Commission consisting of twelve members ⁽¹⁾, set up in 1994, is in charge of assessing the studies. It published two progress reports in June 1995 and June 1996.

Its principal recommendation, to set up the scientific and technical coordination and a research plan, was followed by a letter from the Ministry (December 27, 1995) organising the management of the R&D and by the publication of a research programme.

Among these technical recommendations, the Commission requested that thought concerning the order of precedence to be assigned to the long-lived radionuclides to be separated and transmuted be continued and that quantified goals be defined. It also requested that the studies and research be organised so as distinguish between the short and medium-term options concerning already industrial systems, or those undergoing industrialisation, and the longer-term options which are based on the overall foreseeable innovative systems. Lastly, it considered scenario studies and their contribution in the acquisition of highly diversified data bearing on radionuclide inventories, flows of material, economics, safety, etc. These studies are necessary to assess the partitioning-transmutation systems that result in particularly complex schemes.

Deputy Christian Bataille (Member of Parliament) published a report on the "evolution of research on the management of high level nuclear waste", on behalf of the French Office of Scientific and Technological Choices. He recalled the genesis and the content of the 1991 law, underlining its beneficial effect in calming down the discussion, indicated that the research on the transmutation of radionuclides, the underground disposal of waste and the long-duration interim storage of wastes, is progressing most satisfactorily. In conclusion, he noted that no domain, even technical in appearance, is outside the realm of democracy and that today, the law-maker's will is done. However, the change in the European economic context has modified the picture and he recommends adapting to the postponed reprocessing strategy by promoting new research on the possibilities of long-term surface interim storage as well as on the possibilities of direct disposal of non-reprocessed assemblies.

The exposé below gives the state-of-the-art on the research programme concerning partitioning-transmutation carried out by CEA in collaboration with those "directly concerned by the law" (EDF, COGEMA, CNRS, ANDRA, ...), and replies to two questions:

- What can be done in partitioning-transmutation? At what cost? With what timescale?
- How can long-term gains be qualified and quantified? Are there short-term disadvantages?

⁽¹⁾ The Commission consists of six people (including at least two international experts) appointed by the French National Assembly and by the Senate, and proposed by the French Office for Parliamentary Scientific and Technological Choices, two people appointed by the Government and proposed by the Conseil Supérieur de la Sûreté et de l'Information Nucléaire (Higher Council for Nuclear Safety and Information), four scientific experts appointed by the Government and proposed by the French Academy of Sciences.

THE ASSETS AND LIMITATIONS OF PARTITIONING-TRANSMUTATION

Partitioning-transmutation fulfills the desire to transform long-lived radioisotopes (>31 years) into short-lived radioisotopes or stable isotopes which could then be managed by confinement and natural decay over periods of some hundreds of years.

It is not planned for short-lived radionuclides (< 31 years) for which confinement pending decay is clearly preferable and already implemented, for instance, in the Aube Storage Centre (CSA).

Both operations, partitioning and transmutation, call upon different types of techniques the limitations of which should be clearly understood.

1. Assets and Limits of partitioning-transmutation

1.1. Partitioning: Assets and Limits

Radionuclides are found in non-reprocessed spent fuels, in type B wastes and in glass (table 1, diagram 1).

To achieve transmutations or fissions, the radionuclides must be submitted to a sustained neutronic flux. These conditions lead to the use of ceramic-type materials able to withstand high temperatures, thermal flux and mechanical constraints. It is therefore necessary to have concentrated and purified radionuclides. (The ideal would be an isotopic purity). With the removal of the long-lived radionuclides from the wastes as a goal, the partitioning must be performed with the highest possible efficiency before proceeding to the purification, concentration and production of the ceramics.

The type B wastes represent large volumes in which the radionuclides are diluted. They must be decontaminated to a point where they can be stored on the surface. The feasibility of such operations depends on the type of waste (concrete, metals, etc.) and on the nature of the contamination (labile, fixed etc.). The performance required is beyond our present know-how. On the other hand, the quantities of radionuclides recovered would be small and easy to manage with those from the bulk of the spent fuel.

The spent fuel, when reprocessed, is quasi-totally dissolved. The valuable materials, U and Pu, are separated and purified (with a recovery yield of 99.9%). Iodine and other fission gases (krypton, ...) are discharged. The fission products and minor actinides (Np, Am and Cm) are contained in a so-called "high level effluent" (which contains 98% of the $\beta\gamma$ activity and the α activity exclusive of plutonium). The glass is made from the high level effluent. (Figure 1).

The high level effluent containing most of the radioactivity is the subject of long-lived radionuclide separation studies.

In a first stage, the partitioning-transmutation applies to spent fuels only. It does not apply to type B wastes.

Extraction by solvent techniques are extremely efficient (they are the ones that enable the industrial recovery of uranium and plutonium), on condition that a good solvent is used. The latter must extract the desired element with very high affinity (recovery yield) and good selectivity (without extraction of unwanted species), allow the subsequent recovery of the element (back extraction) and enable the implementation of operations under industrial conditions without generating new waste.

Partitioning is first of all a question of molecular chemistry (discover the functionalities of the molecules of the solvent and tailor the molecule to the extraction conditions), then of chemistry and of chemical engineering (operating conditions).

For neptunium, zirconium and technetium, the research pertains to implementation, as the solvent is well-known (TBP).

For caesium, the calixarenes functionalised by etheroxide chains have been discovered. The development of a molecule for the solvent and process implementation remain to be achieved.

For the co-extraction of actinides and lanthanides, a molecule has just been developed (DMDBTDMA) and the Diamex process is to be defined (chemistry and implementation).

For americium and curium, unsatisfactory solutions exist (very tricky implementation and large secondary wastes) and interesting functionalities are under study (TPTZ and picolinamides). It is hoped that they will be suitable and, if so, the molecules will have to be tailored to the extraction conditions and the implementation made.

Table 2 gives the research status for the partitioning of actinides and fission products.

Concerning partitioning, once the feasibilities have been acquired, performances will be excellent and a preliminary calculation may be made by assuming losses of long-lived radionuclides from 0.1% to 1% in the high level effluent sent to vitrification.

1.2. Assets and limits of transmutation for actinides

Incineration-transmutation (1) is seen by an overlapping of phenomena with the disappearance and the creation of nuclei: the limits of the process therefore have to be clearly defined.

Chemical separation leads to radioisotope mixes. Each one interacts with neutrons in a specific way, which is the only interaction possible today to transmute on a large industrial scale.

Actinides may undergo fissions generating radioactive fission products as well as neutronic captures which produce other actinides. The radioactive decay of actinides spans a very long time and belongs to one of four main families of decay which end with stable isotopes of lead or bismuth.

a) Physical feasibility

Studies to date have shown that it would be possible theoretically, and exclusive of any industrial and economic feasibility studies, to very appreciably reduce the final production of radiotoxic heavy nuclei (plutonium and minor actinides) resulting from the supply of electricity, using fission reactors in a park of reactors that produce and consume these heavy nuclei. Such a park is said to be balanced (in mass and isotopic composition) when it reaches a steady state in which all the radiotoxic heavy nuclei it produced are fissioned.

Various scenarios of multiple recyclings of plutonium alone or with minor actinides in parks consisting of PWRs, RMAs (AMRs?) or FBRs have been studied to strike a balance between production and consumption of actinides, hence a final zero production within the park. (Diagram 2).

These studies tend to show that there are neither outstanding nor even significantly different scenarios for the recycling of plutonium alone (Table 3). They all result in balanced Pu inventories in the cycle, of the same order of magnitude (a few hundred tons). They also show that the in-cycle plutonium inventory is higher for these scenarios than for a PWR open cycle that produces the same amount of energy but discards the plutonium for waste. On the other hand, the amount of plutonium sent to waste in these scenarios depends only on the separate performances of the cycle knowing, moreover, that the production of minor actinides is considerably increased.

(1) The term transmutation is used when the atomic nucleus captures a neutron, and incineration when it fissions.

These studies also indicate that the dynamics of the systems is very slow (about 100 years to reach equilibrium) and that a controlled phasing out of nuclear power must also be provided for to progressively eliminate the plutonium inventory in the cycle. (1% may be reached in more than 100 years).

The same theoretical studies began with the recycling, besides plutonium, of the minor actinides. They started with the study of the reactivity of various reactor cores and the verification of the main safety parameters. Thus the acceptable limit in minor actinides was set at 5% in FBRs and 1 to 2% in PWRs when the minor actinide is homogeneously distributed within the UO₂-PuO₂ mixture in the ceramic fuel. These cores result in actinide incineration performances of 18 to 27% in FBRs and from 6 to 9% in PWRs at standard burn-ups and allow a positive balance in the consumption versus the production of minor actinides (Table 4). Multiple recycling scenarios are under study. It seems that the same type of results would be obtained as with the recycling of Pu alone, slightly more contrasted depending on the reactors. The study with CAPRA reactors is partly completed (Table 5).

The management of curium is difficult owing to its strong neutron and gamma emission. Interim storage may be envisaged (100 years) and then transmute the plutonium from decay or accept to send it to waste pending very strongly reduced production. This could be obtained by separating the americium and by its single-recycling once only in targets. (Then an americium destruction rate by fission higher than 90% is required).

The manufacture of plutonium fuels has already reached the industrial level, and their in-reactor performances mastered. The fuels also containing neptunium raise little problem (an experimental assembly was produced for SPX). On the other hand, the americium targets require further fundamental research on the chemical nature of the americium combination (AmO₂, AmO_{2-x}, Am₂O₃), and of the inert matrix able to behave properly under irradiation (choice of the material, Al₂O₃? MgO?...). It is also necessary to improve the behaviour of the cladding under long irradiation and very great deformation of the ceramics. Table 6 recapitulates the status of the research in progress.

Furthermore, innovative options for transmutation such as hybrid systems based on accelerators are being considered. Their potentialities remain to be analysed.

b) Technical and technological feasibility

Several scenarios should be selected, chosen for their technical and industrial credibility and the broad range of choices offered to decision-makers, and technical work should be performed on the feasibility of reactors, the feasibility of the cycle, the dynamics of the systems, the inventories per cycle and the wastes, and the consequences on the risks and costs. Unsteady state periods will deserve special attention.

1.3. Assets and limitations of transmutation for fission products

Long-lived fission products can be transmuted by neutron capture only. Therefore, a system must be used that produces a large available excess of neutrons. This is why fission reactors (in the theoretical parks mentioned above) are not very efficient for transmuting long-lived fission products.

Each fission product must be studied individually to assess the technical feasibility of its transmutation in reactors or in innovative dedicated systems. Sensitive points appear: the making of thermally stable iodine targets able to withstand the irradiation, the isotopic separation of caesium (without this separation, caesium 133 and 134 generate more caesium 135 than is transmuted, without mentioning the occurrence of the highly radioactive caesium 137), the recovery of carbon 14 which is an activation product of nitrogen 15 and oxygen 17 (can it be separated?)... One case is favourable: that of technetium 99. The others have not yet been studied.

2. Conclusion and prospects

Partitioning-transmutation can be considered from the technical point of view, in a dual approach at the medium and long-term.

At medium term, it is theoretically possible to design nuclear reactor parks that fission all the transuranic heavy nuclei they produce.

The main sensitive points identified today are actinide-lanthanide separation, making americium targets and managing curium.

Major technical changes are necessary, in reactors, reprocessing and fuel fabrication.

These points make up the essential part of the technical research programme.

This management of materials should be performed on a large scale and on an extended timescale. It will not eliminate the "high level and long-lived" wastes as the long-lived fission products and the actinide losses from the cycle (~ 0.1%) will remain in the glass. Type B wastes could be increased in proportion to the flows processed. Furthermore, the controlled phasing out of nuclear must be taken into account.

Several studies of complete scenarios over time, inventory of material, wastes, risks and costs according to the service rendered will be useful to the 2006 decisions and are included in the research programme.

For the long-term, innovative techniques may be required. They could include new hybrid reactor systems allowing to incinerate the capturing only isotopes as well. Their potentiality and the whole of the associated cycle remain to be studied.

A rather fundamental assessment-oriented research programme has been launched.

In any case, type B wastes will remain. Endeavours to rationalise and reduce the volumes are being made in the framework of current reprocessing (Pu partitioning).

The Puretex research programme deals with this subject.

HOW CAN THE ADVANTAGES AND DISADVANTAGES OF PARTITIONING-TRANSMUTATION BE QUALIFIED AND QUANTIFIED?

Transmutation is a complex operation, easier to contemplate today for plutonium than for minor actinides or fission products. It meets the desire to get rid of wastes by destroying them, but it does have limitations. Since it concerns only separated radionuclides and refined products, it cannot not deal with type B wastes.

It does, however, enable the composition of future "high level and long-lived" wastes to be changed by modifying the inventory of the long-lived radionuclides they contain. So it is a technical step in the management of wastes, upstream from their production, and which cannot be assessed independently.

1. Can partitioning-transmutation complement geological disposal?

Projects for disposing of radioactive wastes in geological formations aim at isolating the radionuclides from man and the environment for a long enough time for their impact to have decreased below the regulatory limit of 0.25 mSv/year. The performance of the multibarrier confinement system (package, engineered barrier and geological formation) is assessed by means of safety models taking into account:

- the behaviour of the waste packages,
- the transfer rate of the various radionuclides through the engineered barrier,
- the transfer rate of the various radionuclides in the geosphere,
- the transfer paths and times of the various radionuclides within the biosphere,
- the effects of the various radionuclides on man.

The different confinement barriers allow to limit the flux of radionuclides at the outlet and hence their impact on man. The safety assessment allows this impact to be assessed for each radionuclide versus time, and hence to identify and classify the radionuclides that are important for safety. In particular, it allows to identify those that would be at the origin of a relatively greater impact, linked either to their properties, or to the uncertainty concerning their behaviour. These calculations are performed right from the design phase, taking the conditions offered by the site into account.

In the event the impact of some radionuclides from storage in a given site were too great in view of regulatory requirements, means must be found to reduce it. If their management involved technical conditions that are difficult to achieve, other processes must be considered. Several paths may then be envisaged, including:

- a reduction at the source of the wastes disposed of, corresponding to a reduction in the radionuclide inventory. This is the partitioning followed by transmutation system,
- partitioning for special conditioning,
- adapting the performance of the engineered barrier to meet the safety requirement,
- adding an extra barrier, of the over-packing type or similar.

Each of these systems must be examined and assessed with respect to the safety objectives and the technical means to reach them.

Some orientations may already be provided based on generic type exercises, including the international exercises. They indicate that, in a normal evolution scenario:

- the actinides are well contained by the multibarrier system. Owing to their very low solubility in a reducing medium, they are immobilised by precipitation either in the barriers or within the natural system,
- the only significantly predominant impact could be due to mobile fission products. Iodine is often mentioned, and to a lesser extent caesium. Elements for which the data on behaviour in a natural medium are still uncertain are also mentioned: technetium, palladium, niobium and selenium.

A specific request concerning partitioning and transmutation originating from disposal studies can therefore not be considered before the end of 1997, namely in the framework of the first choice of concept, once the inventory per radionuclide has been specified, the long-term behaviour of the packages described and the specifications laid down.

2. What does partitioning-transmutation contribute as a way to reduce the inventory of long-lived radionuclides in waste?

It may first be considered that P&T fits into a simple rationale to reduce the inventory of long-lived radionuclides sent to waste to potentially reduce the dangers that future generations may be subjected to. In that case, it results only from a subjective need to reduce "potential" dangers independently of any waste management scenario.

The analysis of the concentration of radioisotopes in a spent fuel, of their specific radioactivity and of their dose factor when ingested or inhaled has shown that the "radiotoxic inventory" after 1000 years was linked only to the actinides a thousandfold to ten thousandfold more than to the long-lived fission products. Plutonium represents 90% of the inventory, minor actinides 10%. Plutonium and minor actinides are therefore the priority challenge (Figure 2).

Let us recall that the "radiotoxic inventory" is only a global indicator. It is obtained by weighing each element in the inventory (its concentration) by a coefficient which is strictly valid only in the event of inhalation or ingestion in small doses.

Reducing the inventory of plutonium and minor actinides in wastes obliges global scenarios to be taken into account when they are recycled in a reactor park.

These theoretical scenarios compared with the once-through scenario where the spent fuel is considered a waste, brings out the following conclusions:

In the case of multirecycling plutonium alone, the radiotoxic inventory of the wastes is only reduced by a factor less than 10 (from 2 to 5 depending on the reactor type), since 10% of the plutonium is transmuted into americium and curium (Figure 3 and 4 give an example for Pu recycling in PWRs).

In the case of multirecycling of plutonium and minor actinides, the inventory gain in wastes will then practically depend only on the partitioning performances.

a P&T strategy in a reactor park really makes sense only if it is mastered from beginning to end: reaching a steady state in the park, operating this balanced park over the desired period of time, programmed phasing out necessary to eliminate the inventory in the cycle and putting an end to the nuclear programme. Any brusque interruption in the sequencing of these phases would heavily penalise, or even annihilate the expected gains in the tonnage to be disposed of.

the establishment, operating and programmed phase-out times for nuclear in a P&T strategy is counted in centuries. The problem of the availability of the foreseeable uranium resource in a PWR park will then be acutely felt for carrying out such a strategy. In this regard, the position of FBRs is much more favourable in all respects: sufficient uranium resources, proven physical feasibility of multirecycling, lower production of minor actinides, etc.

The inventory of fission products in wastes can hardly be reduced using fission reactors and dedicated, potentially more efficient, innovative systems must be considered. However their feasibility remains to be proven.

CONCLUSION

Partitioning-transmutation applied to the management of materials is an operation that could modify the composition of the wastes and act on the long-lived radionuclide inventory (especially actinides). We do not know today how to assess the gain of such operations, or the disadvantages resulting from a service rendered, namely the TWhe of electricity produced.

This step will have to be included in nuclear power evolution and waste management scenarios to bring out more significant parameters of the pros and cons, for example the short-term radiologic impact on the workers, the impact that could be produced in the future by geological disposal, the costs, the consumption of raw material resources, etc.

Typical scenarios could perhaps suffice to bring out easier to use assessment criteria by showing "classes" of radionuclides (fissile materials, γ n emitters, mobile elements, very low radioactive β emitters, etc.).

However, the difficulty in short-term and long-term risk intercomparisons will remain.

Acknowledgments:

The results quoted were obtained at CEA and through a very large number of research partners. In France: EDF, COGEMA, CNRS. In Europe: the European Communities, TUI, PSI (Switzerland), ECN (Holland), FZK (Germany). In Japan: PNC, JAERI. In Russia: Minatom, IPPE, IPC.

May they all be thanked.

TABLE 1

**LONG LIVED NUCLIDE INVENTORY IN 2010,
ACCORDING TO EDF'S PLAN (GLOBAL 1995)**

FUELS:		WASTES:	
PWR operation buffer (UOX)	= 8 500 mtHM	GLASSES	= 2 800 m ³
PWR in storage (UOX)	= 3 440 mtHM	Type B	= 70 000 m ³
PWR in storage (MOX)	= 1 900 mtHM		

ACTINIDES (metric tons)

	Pu	Np	Am	Cm
In glass and type B waste	0.15	7.6	7.1	0.9
In stored fuels and operation buffer	223 (1)	6.9	17	2.3

LONG LIVED FISSION PRODUCTS (metric tons)

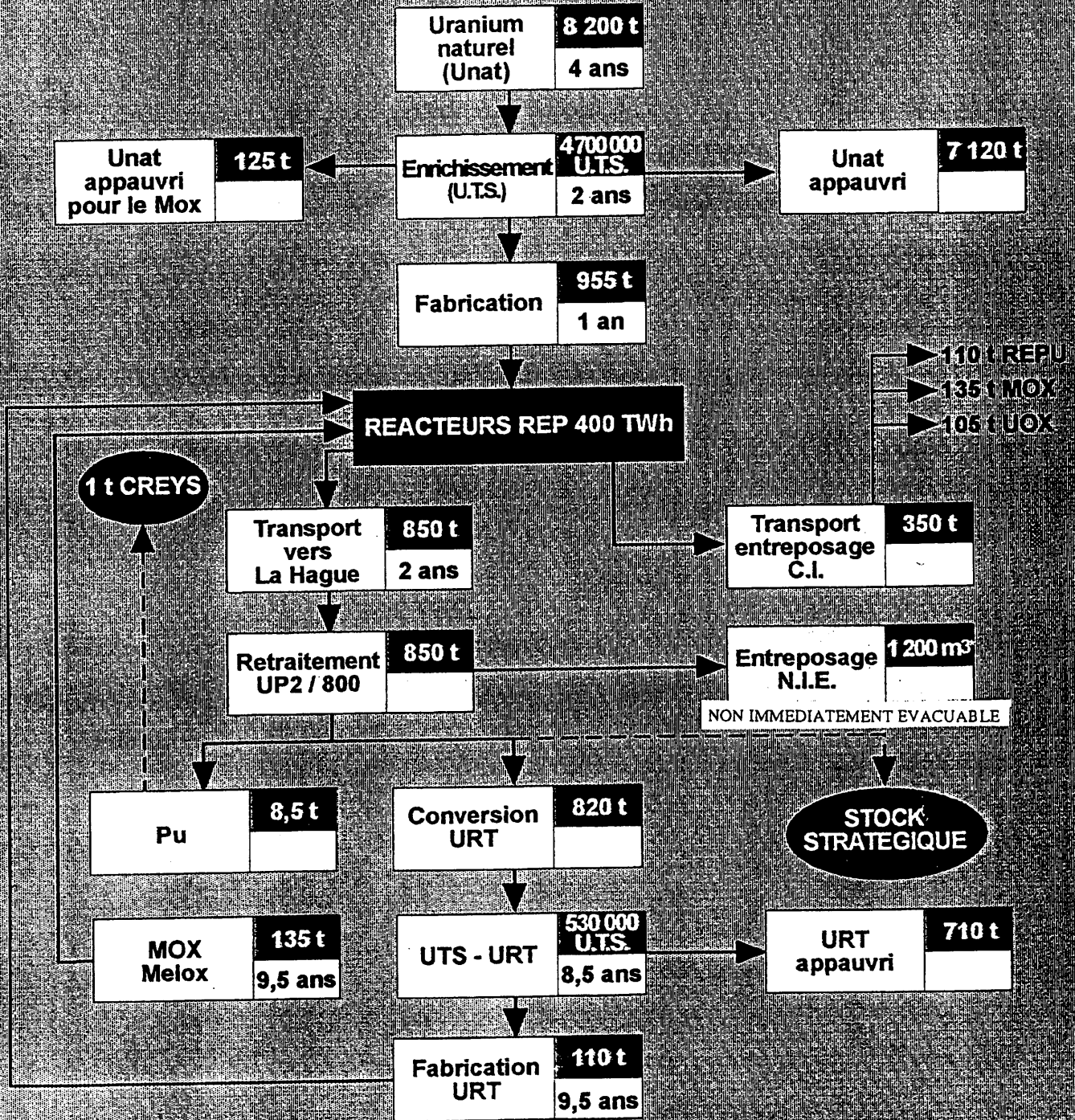
	⁷⁹ Se	⁹³ Zr	⁹⁹ Tc	¹⁰⁷ Pd	¹²⁶ Sn	¹²⁹ I	¹³⁵ Cs
In glass and type B waste	0.01	9.7	13.7	3.7	0.4	---	6.3(2)
In stored fuels and operation buffer	0.01	11	13.8	4.4	0.4	3	7.2

(1) Including 100 tons of Pu in MOX fuel assemblies

(2) ¹³⁵Cs = 6.3 tons for Cs total = 44 tons



LES FLUX DANS LE CYCLE DU COMBUSTIBLE NUCLEAIRE



étape du cycle	flux de matière URT (t _{ML})	date de l'opération avant chargement ou après déchargement
----------------	--	--

* dans une hypothèse favorable de conditionnement pour entreposage à long terme, les 850 t d'assemblages irradiés occuperaient 1 700 m³.

** Pour les besoins de la présentation, on a retenu un flux de retraitement saturant la capacité nominale d'UP2 et l'égalité des flux de retraitement/MOX/URT sans prendre en compte les besoins de Creys.

FIGURE 1 : BEHAVIOUR OF LONG-LIVED ELEMENTS IN THE PUREX PROCESS

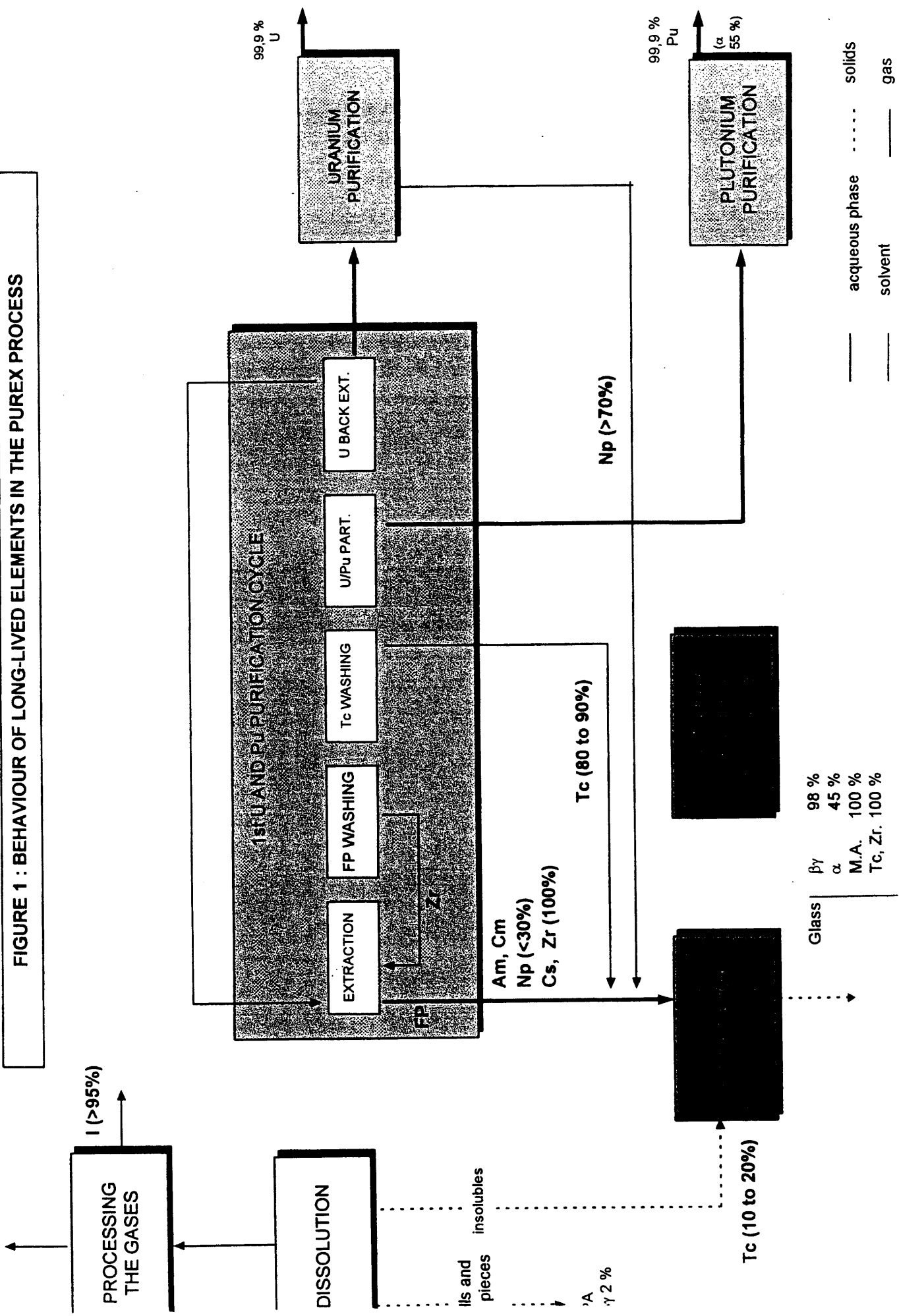


TABLE 2

RESEARCH STATUS ON ACTINIDE AND F.P. PARTITIONING

	Basic R & D	Process Development	Process Industrialisation
U/Pu partitioning			
Np partitioning		X > 99 %	X 95 %
Am/Cm partitioning			
Diamex		X	
Ac/Ln	X		
Oxidised Am partitioning	X		
Tc partitioning	X insoluble fraction		X soluble fraction
I partitioning			X 95 %
Zr partitioning		X	
Cs partitioning	X		
Pd, Se, Sn partitioning			
X : Present status of research			

Once the feasibility is demonstrated, performances will be excellent and 99 to 99,9 % of the radionuclides should be separated.

SCENARIOS OF MULTIRECYCLING OF PLUTONIUM ALONE
OR WITH MINOR ACTINIDES

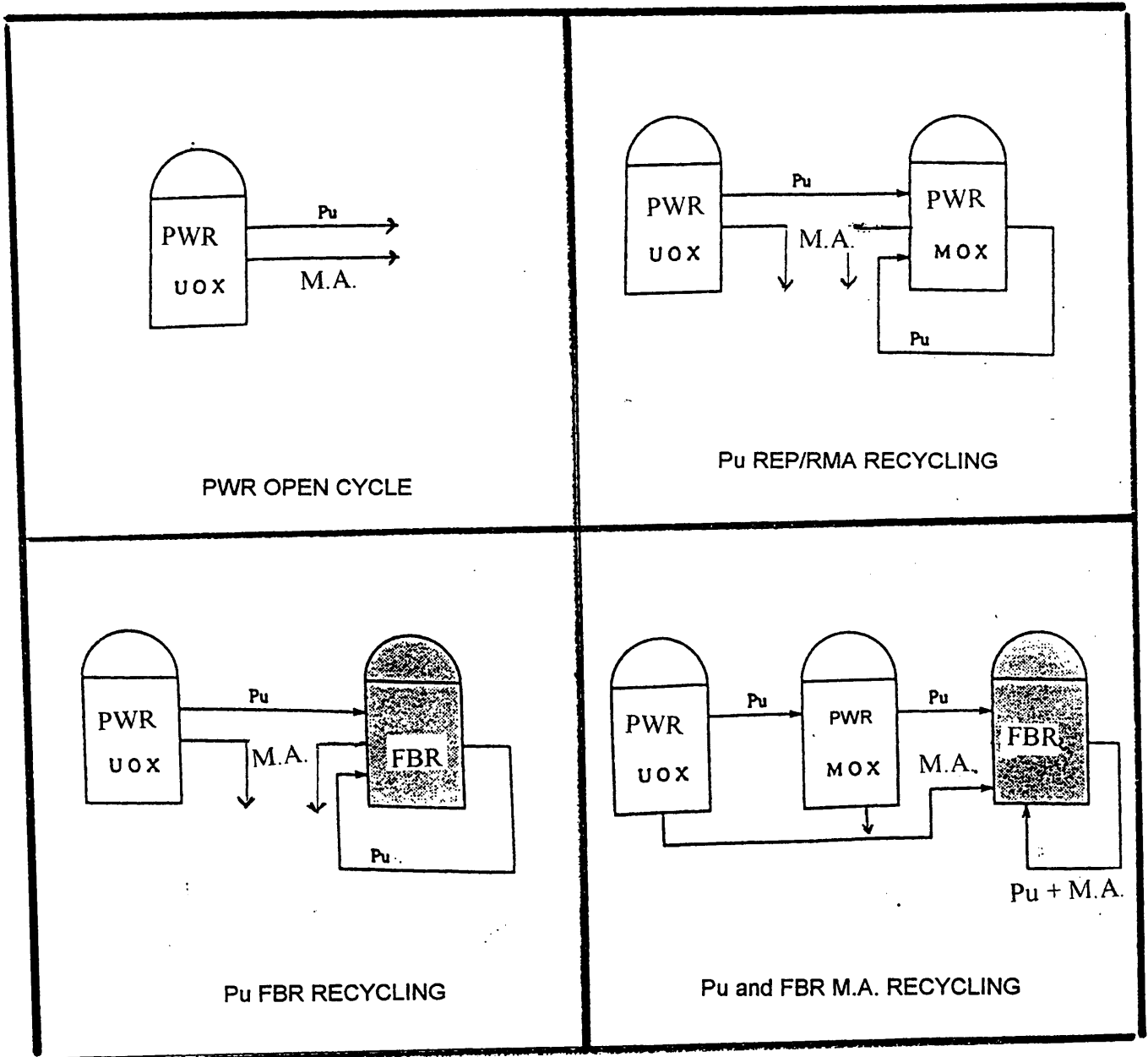


DIAGRAM 2

TABLE 3

MULTIRECYCLING OF PLUTONIUM ONLY
 STEADY STATE CONDITIONS
 60 GWe - 400 TWh

Reactor	N4 MOX heterogeneous	N4 MOX homogeneous	RMA heterogeneous	RMA homogeneous	CAPRA	N4 UOX
Moderation ratio	2	2	4	4	---	2
U, associated to Pu (%235U)	2.8%	3.8%	0.25%	3.8%	0.19%	4.5%
Pu concentration (%)	9.6%	2%	18%	1.2%	54%	0
Cycle inventory in tons of Pu	400	200	390	112	310	35
Pu in wastes (kg/year)	28	16	31	10	37	11600
MA in wastes (kg/year)	3600	3250	3450	2500	2800	1520

TABLE 4

PHYSICAL FEASIBILITY OF RECYCLING MINOR ACTINIDES IN REACTOR

	Maximum concentration in M.A.	Transmutation ratio
PWR N ₄ or RMA (47 GWJt ⁻¹)	1 to 2 %	6 to 9 %
FBR (EFR type) (120 GWJt ⁻¹)	5 %	18 to 27 %

Homogeneous recycling (minor actinide dispersed in a standard fuel).

Results : 3 to 15 kg/TWhe transformed depending upon reactor type and recycling conditions against 3 kg/TWhe produced in a PWR (UOX) at 33.000 MWdt⁻¹

TABLE 5

EXAMPLES OF INVENTORY REDUCTION (Pu and M.A.)
IN WASTES (Pu and M.A. RECYCLING)

400 TWhe/year

<u>PWR open cycle (UOX)</u>	<u>Steady state park with FBRs (CAPRA)</u>
<ul style="list-style-type: none"> • Yearly uprisings: Pu 12 mt/year M.A. 1.3 mt/year • Yearly amounts to waste: Pu 12 mt/year M.A. 1.3 mt/year • In-cycle inventory~In-reactor inventory Pu 34.8 mt M.A. 3.9 mt 	<ul style="list-style-type: none"> • Yearly uprisings: Pu ~ 0 M.A. ~ 0 • Yearly amounts to wastes: Pu ~ 0.1 mt/year M.A. ~ 0.1 mt/year depending upon separation efficiency • In-cycle inventory ~ In-reactor and in-plant inventory Pu ~ 460 mt (170 tons in reactors) M.A. ~ 120 mt (50 tons in reactors)

A STEADY STATE FBR PARK VERSUS A PWR OPEN CYCLE

	Over 10 years	Over 100 years
Inventory in wastes	: 70	: 70
Total inventory (cycle and wastes)	X 3.5	: 2.2

TABLE 6

RESEARCH STATUS ON THE DEFINITION OF FUELS AND TARGETS FOR
TRANSMUTATION

	Basic R&D	Process development	Process industrialisation
FUELS			
UO ₂ or PuO ₂ + Np + Am + Cm		X X	X
TARGETS			
Am targets Cm targets Tc targets	X	X	
X: Present status of research			

FIGURE 2 : Radiotoxic inventory of one ton of UOX1 fuel

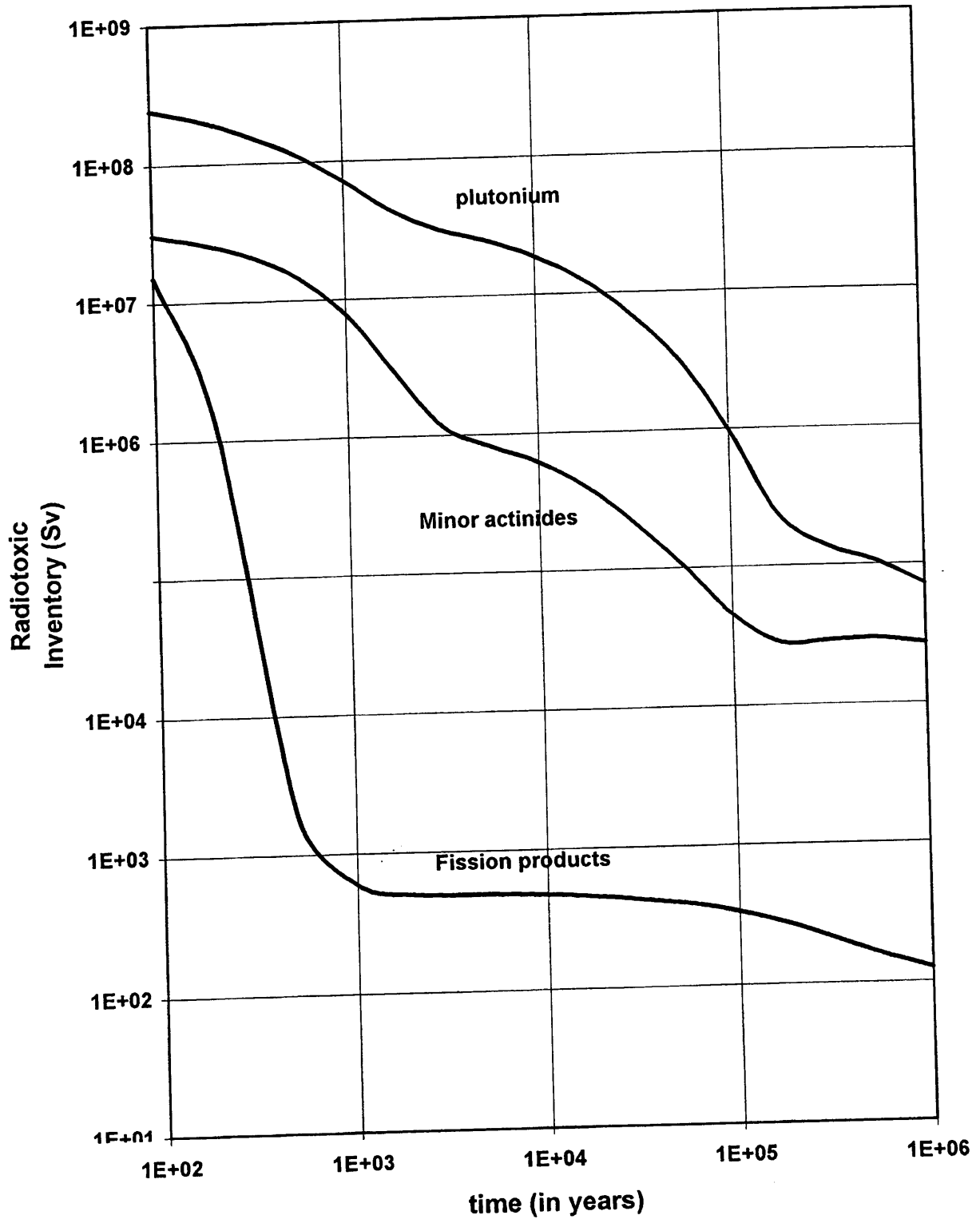


FIGURE 3: Contribution of the various initial actinides to the radiotoxic inventory of wastes
(UOX1 fuel case)

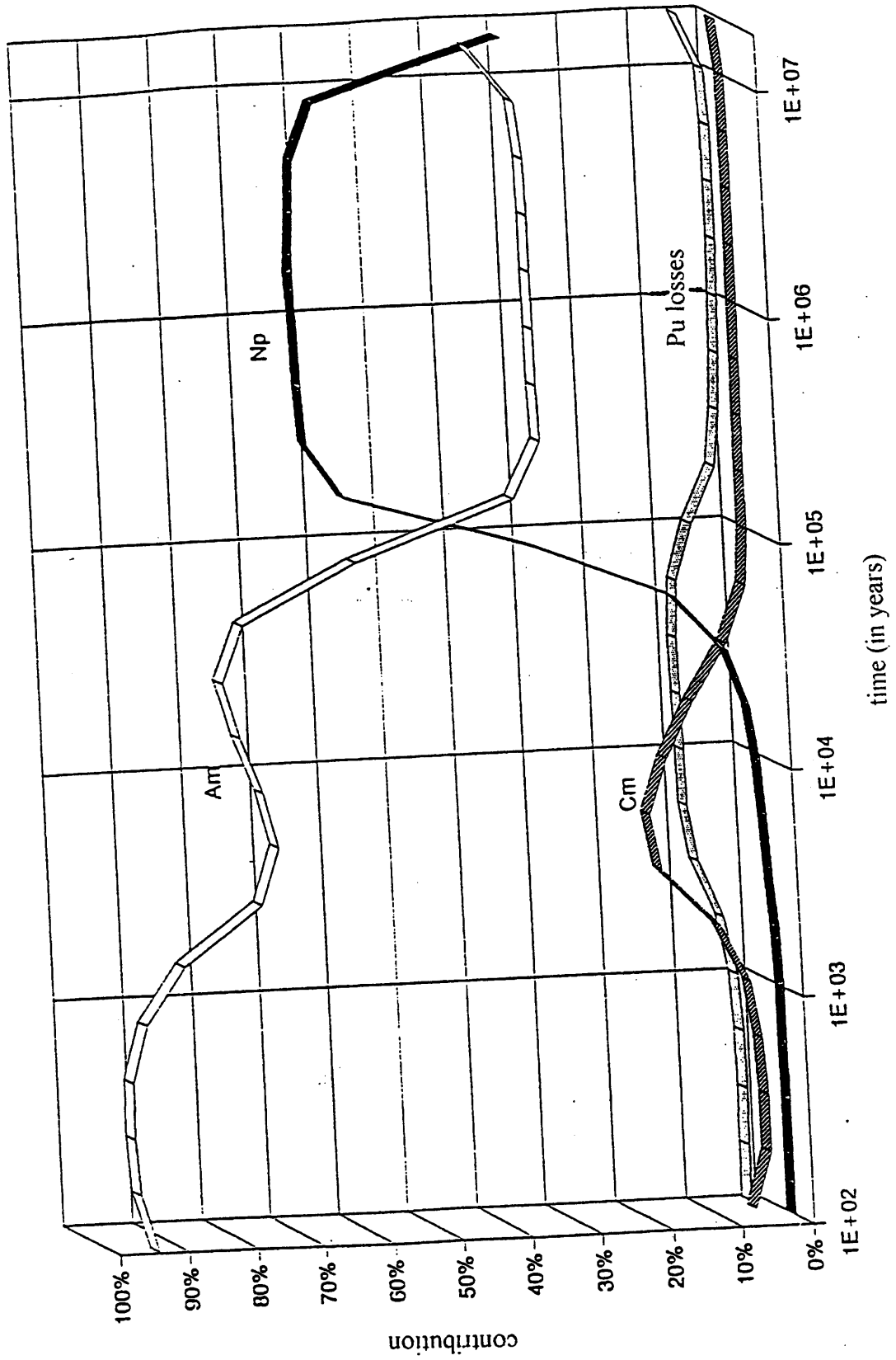
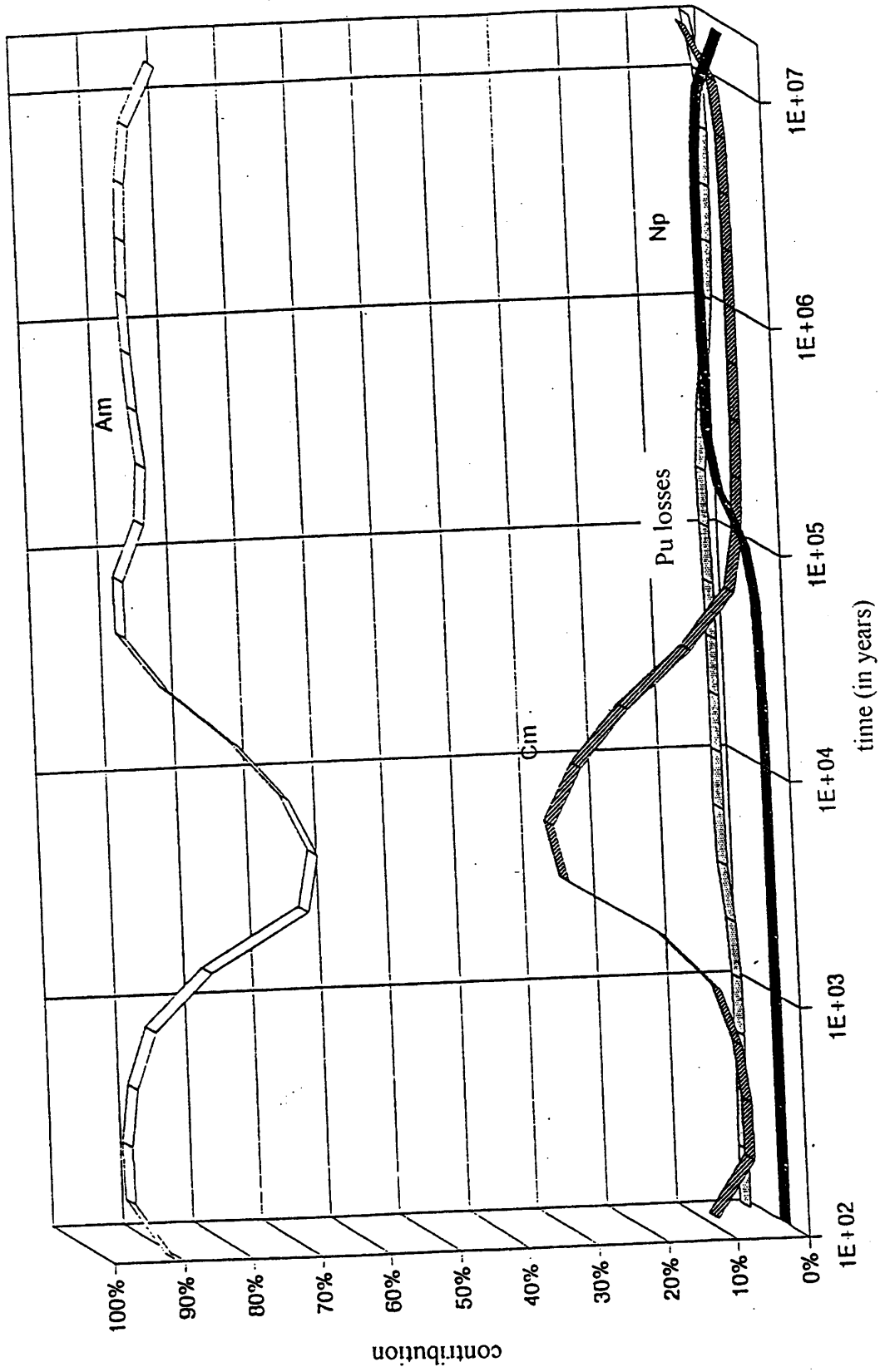


FIGURE 4: Contribution of the various initial actinides to the radiotoxic inventory of wastes (MOX fuel case)



**PARTITIONING AND TRANSMUTATION PROGRAM
"OMEGA" AT JAERI**

T. Mukaiyama, M. Kubota, T. Takizuka,
Y. Suzuki, T. Ogawa, T. Osugi, and M. Mizumoto

JAPAN ATOMIC ENERGY RESEARCH INSTITUTE
Tokai-mura, Ibaraki-ken, 319-11 Japan

ABSTRACT

During the last two decades, JAERI has been carrying out the partitioning and transmutation program in the following areas: (a) development of the four-group partitioning process, (b) design study of the transmutation systems, one with actinide burner reactor, the other with an accelerator-driven subcritical reactor, (c) development of an intense proton accelerator, (d) development of nitride fuel manufacturing and pyrochemical reprocessing, and (e) basic research for supporting the development of transmutation systems. Activities and recent achievements are overviewed.

JAERI is about to launch the Neutron Science Project which aims at bringing scientific and technological innovation for the 21st century in the fields of basic science and nuclear technology using neutrons. The accelerator-driven transmutation system study and the development of an intense proton accelerator are also under way as important parts of this project.

I. INTRODUCTION

Since the mid 1970s, JAERI has been developing a partitioning process of high-level waste (HLW) and a concept of a dedicated transmutation system of minor actinides (MAs) as an effective and efficient measure to alleviate the long-term burden of nuclear energy which may arise from the management of HLW. During these studies, the double strata fuel-cycle concept was developed. JAERI's activities cover the following areas of the partitioning and transmutation: (a) development of the four-group partitioning process, (b) design study of the actinide burner reactor and the accelerator-driven hybrid system, (c) development of an intense proton accelerator, (d) development of nitride fuel cycle technologies, and (e) basic research for supporting the development of transmutation systems.⁽¹⁾

In the course of planning of an intense proton accelerator for transmutation, we recognized that neutron scattering researchers need intense neutron beams for basic science. JAERI is planning to launch the Neutron Science Project which aims at accomplishing the scientific and technological innovation using neutrons for the 21st century. An intense proton accelerator will be constructed for dual duty as a neutron producer for transmutation and for basic science including neutron scattering.

II. DOUBLE STRATA FUEL CYCLE

JAERI has been proposing the concept of the double strata fuel cycle consisting of the power reactor fuel cycle and the Partitioning-Transmutation (P-T) cycle.⁽²⁾ In this scenario, a reprocessing plant, a partitioning plant and a transmutation plant will be co-located in one site. This configuration of plants composes a high level radioactive waste (HLW) management park.

The concept of the double strata fuel cycle is illustrated in Fig. 1. The final HLW from this fuel cycle contains only short-lived fission products. The separate treatment or the isolation of MAs from the commercial cycle is preferable from two reasons: (a) complete recovering and transmutation of MAs and long-lived fission products are possible, and (b) MAs are strong neutron emitters, and, thus, MA recycling through the conventional fuel cycle may cause problems in the radiation shielding of the fuel cycle facilities. This may cause the cost of electricity generation to increase.⁽³⁾

III. DEVELOPMENT OF FOUR-GROUP PARTITIONING PROCESS^(4,5)

Firstly, a partitioning process was developed for separating elements in HLW into three groups, namely, transuranium elements (TRU), Sr-Cs, and others. The process consists of three steps: (a) the solvent extraction of U and Pu with tributylphosphate (TBP), (b) the solvent extraction of Am and Cm with diisodecylphosphoric acid (DIDPA), and (c) the adsorption of Sr and Cs with inorganic ion exchangers. The process was demonstrated with actual HLW and more than 99.99% of the Am and Cm were extracted with DIDPA.

Later, a four-group partitioning process has been developed in which one step for separating the Tc-PGM group was developed in addition to the three-group separation. Effective methods for separating TRU (especially Np) and Tc have been developed.

(a) Np Separation

Neptunium, which is dominant in HLW, is the most difficult actinide element to be extracted with general organic solvents. A new process was developed in which more than 99.97% of penta-valent Np was extracted when hydrogen peroxide was fed to a level to compensate for its decomposition in the DIDPA extraction process.

(b) Am and Cm Separation from rare earths

Selective stripping of Am and Cm from DIDPA with the complex agent DTPA is being studied for their separation from rare earths (RE). Batch experiments showed that the separation factor between Am and RE (ratio of distribution ratios) is larger than 10. Experiments are now in progress to find the optimum process conditions.

(c) Tc Separation

Two methods have been developed to separate Tc, precipitation by denitrating HLW and adsorption with active carbon. More than 95% of Tc in a simulated HLW was recovered as precipitate by denitration of HLW. An active carbon column was used for the quantitative adsorption of Tc from a 0.5 M nitric acid solution. Desorption of Tc from the column was achieved quantitatively by the use of an alkaline thiocyanate solution as eluant.

The four-group partitioning process was developed as shown in Fig. 2. This process is to be tested with actual HLW at NUCEF (Nuclear Fuel Cycle Safety Engineering Facility).⁽⁶⁾ Hot operation with actual HLW is scheduled in 1998.

IV. DESIGN STUDY OF THE TRANSMUTATION SYSTEMS

A transmutation system with a very hard neutron energy spectrum and high neutron flux would be very efficient and effective for MA transmutation. The concepts of MA burner reactors (ABR: Actinide Burner Reactor) and a proton accelerator-driven MA transmutation system have been developed at JAERI. When a dedicated transmutation system becomes available, the scheme of an entire fuel cycle will be a strata structure fuel cycle.

IV.1. MA BURNER REACTORS (ABR: ACTINIDE BURNER REACTOR)^(7,8)

Two types of ABRs are designed at JAERI. Fuel material of these ABRs is MA-U nitride mixture. One type is a lead-cooled pin fuel ABR (L-ABR) and the other is a He-cooled particle fuel ABR (P-ABR). Nitride was selected as the fuel material of these ABRs because of its good thermal property. The other advantage of nitride fuel is that it can be processed with the pyrochemical reprocessing, and, hence, the fuel cycle facilities can be very compact and cost effective. The fuel concept of P-ABR is shown in Fig. 3. The reactor core design parameters of these ABRs are given in Table 1. In these ABRs, neutron energy spectrum is very hard and the core-averaged neutron energy is around 720 keV. These hard neutron spectra are very effective for direct fission of MAs which has fission threshold above 600 keV. The MA burnup in the ABRs of 1 GW thermal output is 190 to 200 kg per year.

IV.2. ACCELERATOR-DRIVEN TRANSMUTATION SYSTEM⁽⁹⁾

Two types of accelerator-driven system concepts are being studied; namely, a solid system a molten-salt system. Either system utilizes the hard neutron spectrum of spallation neutrons to transmute MAs efficiently by fission.

(a) Concept of solid target/core system

An accelerator injects proton beam through a beam window into the solid tungsten target located at the center of the target/blanket. Surrounding the target is the subcritical blanket loaded with actinide alloy fuel. Spallation neutrons emitted from the target induce fission in the actinide blanket region. The schematic diagram of the proposed transmutation system concept is shown in Fig. 4. With a 1.5 GeV, 39 mA proton beam, a sodium-cooled solid target/core having k_{eff} of 0.89 produces 820 MW of thermal power. Assuming a load factor of 80%, MA burnup is approximately 250 kg/y, or 8% of inventory per year.

(b) Concept of molten-salt target/core system

The molten salt acts both as fuel and as target material, and also serves as coolant. Its main advantage is the capability of continuous on-line processing of MAs and reaction products. Chloride salt is chosen based on the consideration about actinide solubility and nuclear characteristics. The molten-salt target/core with k_{eff} of 0.92 produces 800 MW thermal power with a 1.5 GeV, 25 mA proton beam. Assuming a load factor of 80%, the MA burnup is approximately 250 kg/y, or 4.6% of inventory per year.

The support factor, which is defined as the number of power reactor units whose MAs are transmuted by one unit of a transmutation system, is about 10 to 15 for dedicated transmutation systems, while the support factor of FBR proposed so far is between 4 to 6.

V. TRU NITRIDE FUEL AND FUEL CYCLE ^(10, 11)

In a double-strata concept, MAs from the commercial fuel cycle flow into the second-stratum of transmutation ("dedicated actinide-burner") cycle. MAs are concentrated and confined in the second stratum, exiting only after being converted to fission products. Considering the inherent difficulty in handling MAs, an innovative approach is required in designing a fuel cycle system for the actinide burning.

Concepts of dense fuel cycles for the second stratum (Fig. 5) have been proposed, where high atom densities of the actinides are maintained throughout the whole cycle, and the system volume and envelope are minimized. JAERI is studying the feasibility of employing the nitride fuel and pyrochemical reprocessing. The favorable thermal properties of the nitride fuels that make full utilization of a cold-fuel concept possible are: (a) lower fuel temperatures and hence lower fission gas release, (b) a thinner cladding to achieve a harder neutron spectrum, and (c) a relatively large Doppler effect in the over-power events.

Efforts are directed to technical developments and fundamental property studies of (a) the sol-gel process to obtain nitride microspheres from the actinide nitrate solution, and (b) the electrorefining process of the nitride fuel with a LiCl-KCl-AnCl₃ (An: actinides) melt.

(a) Nitride fabrication from actinide salts

Actinide nitrate solution from the partitioning of HLW can be solidified to a ceramic form by an internal gelation method. A droplet of the actinide nitrate solution with a carbon suspension turns into a solid mixture of oxide (hydroxide) and carbon in a form of microspheres. A microwave gelation apparatus has been developed and tested at JAERI. The (oxide+carbon) microspheres thus obtained are converted to the nitride by a carbothermic synthesis.

The nitride fuels can be used in the form of either pellets or TiN-coated particles. In the particle-fuel concept, the TiN coating consists of both high-density and low-density layers.

(b) Electrorefining of nitrides

In the proposed pyrochemical process, the irradiated nitride fuels are electrorefined in a LiCl-KCl eutectic melt. Like metal fuels, the actinide nitrides would be anodically dissolved. The design of the electrorefiner may be very similar to that for the metal fuels. The recovered metals then have to be converted to nitrides.

Laboratory runs of the fused salt electrolysis of UN have been made. The recovery of uranium metal has been demonstrated. During the electrolysis, the system was purged with purified helium. Conversion of the metal to the nitride has been readily made in liquid Cd with nitrogen cover gas. The reaction products were U₂N₃ and (U, Gd)N.

VI. DEVELOPMENT OF A HIGH-INTENSITY PROTON ACCELERATOR ⁽¹²⁾

A high-intensity proton linear accelerator with a beam power up to about 10 MW has been proposed for basic science and various engineering tests of the transmutation system.

The R&D work has been carried out for the components of the front-end part of the proton accelerator: ion source, radio-frequency quadrupole (RFQ), drift tube linac (DTL), and RF source. In the beam test, a current of 70 mA with a duty factor of 10% has been accelerated from the RFQ at an energy of 2 MeV. A hot test model of the DTL for high-power and high-duty operation has been fabricated and tested.

For the high-energy portion above 100 MeV, superconducting accelerating cavity is studied as a main option. The superconducting linac is expected to have several favorable characteristics for high-intensity accelerator such as shorter length acceleration, large bore radius resulting in low beam losses, and cost effectiveness for construction and operation. The design work for superconducting cavities is in progress in collaboration with the KEK (National Laboratory for High Energy Physics). A test stand with the equipment of cryogenics system, vacuum system, RF system, and cavity processing and cleaning has been prepared to test the physics issues and

fabrication process.

The main accelerator components such as high-current hydrogen-ion source, RFQ, DTL, and RF power source have been constructed and tested. A high brightness (140 mA) hydrogen ion beam has been extracted.

The first 2 MeV beam test with the ion source and RFQ in combination of a single unit of high power RF source was successfully carried out with the peak acceleration current of 70 mA (a duty factor of 10%).⁽¹³⁾

VII. BASIC RESEARCH SUPPORTING THE DEVELOPMENT OF TRANSMUTATION SYSTEMS

To support the above-mentioned activities on transmutation system development, several basic research activities are in progress.

VII.1. NUCLEAR DATA FILE FOR TRANSMUTATION SYSTEM DESIGN STUDY⁽¹⁴⁾

Two types of nuclear data files are being compiled. The JENDL Actinide File is compiled for use in the ABR design study. It contains neutron-induced reaction data for about 90 nuclides from ²⁰⁸Tl to ²⁵⁵Fm. This compilation will be completed by 1997. The JENDL High-Energy File contains the data for protons and neutrons up to a few GeV which are used for the design of the accelerator and for studying an accelerator-driven transmutation system.

VII.2. MEASUREMENT AND EVALUATION OF ACTINIDE NUCLEAR DATA

MA nuclear data are measured for fission-neutron yields, delayed-neutron yields, and fission yields in collaboration with the Oak Ridge National Laboratory (ORNL) and Texas A&M University. Actinide nuclear data in the JENDL File are evaluated using the integral experiments at the fast critical facility FCA.⁽¹⁵⁾

Spallation integral experiments have been carried out to obtain data on nuclide production, to estimate the yields of neutrons and spallation products, and to evaluate the validity of the simulation code system. The 500 MeV booster proton synchrotron facility at the National Laboratory for High Energy Physics (KEK) is used for the activation experiments. In general, the calculation with the codes agree fairly well with the experimental results. There are however some discrepancies in both the nuclide production cross section and the neutron energy spectrum.^(9, 16)

VII.3. FUEL MATERIAL THERMODYNAMIC DATABASE^(10, 11)

In view of the feasibility study of the above dense fuel cycles, thermodynamic data base needs to be expanded in three major areas: actinide alloys and intermetallics, nitrides, and actinide-containing molten salts. In JAERI, a pure substance and solution data base, related to the actinide burning, is being formed as a minor supplement to the existing thermodynamic data base.

VIII. NEUTRON SCIENCE PROJECT OF JAERI

Neutron scattering has achieved some notable successes in recent years, such as unraveling the crystal structures of high-temperature superconductors, and is now exciting a lot of interest among biologists for probing large organic molecules. A limiting factor for neutron scattering is the intensity of the neutron beams. High intensities of neutron beams allow researchers to carry out experiments that would otherwise be impossible. In Europe, the European Spallation Source ESS is under design, and in the USA, the National Spallation Neutron Source (NSNS) will be constructed at ORNL, both with 5 MW proton beam power.

JAERI is preparing to launch the neutron science project next year. The objective of the project is to construct an intense proton accelerator with proton energy of 1.5 GeV, proton beam power of 6 to 8 MW, and research facilities to be dedicated for the neutron science research center of JAERI Tokai. The spallation neutron source has double duty as a neutron source for basic science and as a neutron source for nuclear-energy-related research such as an accelerator-driven transmutation study.

Basic science in this center covers the fields of structural biology for investigating the structure and function of biological molecules such as DNA, advanced material science (e.g., under extreme conditions), high-energy neutron science (e.g., spallation phenomena), nuclear cross-section measurements for transmutation study, heavy-ion science for creating unstable heavy nuclei through spallation, and decay property measurements of extremely-neutron-rich nuclei.

The layout of the proposed facilities and the proposed schedule for construction of facilities are shown in Figs. 6 and 7.

Transmutation study at this facility will cover the two steps of accelerator-driven transmutation system development. The first step will be the feasibility study of the hybrid system concept at low power level of uranium subcritical system. These experiments will check the stable operation of a hybrid system and MA burnup with use of MA foils or pellets for activation measurements or destructive analysis. The data base for designing a spallation target for high-power experiments will be obtained at the second step. In the first step, most of the transmutation experiments will be performed with pulse-mode operation so that proton beams from the accelerator can be shared in time among the other experiments.

Once the feasibility of the hybrid system concept for the MA transmutation is proven, a test reactor facility with 10-30 MW thermal power will be proposed for the second phase of development. Operation of the hybrid system with uranium subcritical system will be tested at this high power level and transmutation capability will be tested using MA target pins. Technical feasibility of spallation target and beam window will be also tested in the second-step experiments.

REFERENCES

- (1) T. Mukaiyama, M. Kubota, T. Takizuka, T. Ogawa, M. Mizumoto, and H. Yoshida: "Partitioning and Transmutation Program "OMEGA" at JAERI, Proc. Int. Conf. on Evaluation of Emerging Nuclear Fuel Cycle Systems," GLOBAL'95", Vol. 1, p. 110 (Versailles, 1995).
- (2) H. Murata and T. Mukaiyama: Atomkern-Energie Kerntechnik, 45, p. 23 (1984).
- (3) T. Mukaiyama: "Importance of the double strata fuel cycle for minor actinide transmutation", Proc. 3rd OECD/NEA Int. Information Exchange Mtg. on P-T, p. 30 (Cadarache, 1994).
- (4) M. Kubota, I. Yamaguchi, Y. Morita, Y. Kondou, K. Shirahashi, I. Yamagishi, and T. Fujiwara: "Development of a Partitioning Process for the Management of High-Level Waste", Proc. Int. Conf. on Emerging Nuclear Systems," GLOBAL'93", Vol. 1, p. 588 (Seattle, 1993).
- (5) Y. Morita *et al.*: "Diisodecylphosphoric acid, DIDPA, as an extractant for transuranium elements", in the poster session "Chemistry and Reprocessing" Proc. Int. Conf. on Evaluation of Emerging Nuclear Fuel Cycle Systems," GLOBAL'95", Vol. 2, p. 1163 (Versailles, 1995).
- (6) K. Tonoike *et al.*: "NUCEF Project and Its Contribution to Emerging Fuel Cycle Back-End", Proc. Int. Conf. on Emerging Nuclear Systems," GLOBAL'93", Vol. 1, p. 555 (Seattle, 1993).
- (7) T. Mukaiyama *et al.*: "Conceptual study of actinide burner reactors", Proc. Int. Reactor Physics Conf., Vol. IV, p. 369 (Jackson Hole, 1988).
- (8) T. Mukaiyama, T. Ogawa, and Y. Gunji: "Minor actinide burner reactor and influence of transmutation on fuel cycle facilities", Proc. Technical Committee Meeting on Safety and Environmental Aspects of P-T (1993), p. 105 (IAEA-TECDOC-783, 1995).
- (9) T. Takizuka *et al.*: "Research and development on proton accelerator-based transmutation of nuclear waste", Int. Conf. on Evaluation of Emerging Nuclear Fuel Cycle Systems," GLOBAL'95" Vol. 1, p. 489 (Versailles, 1995).
- (10) T. Ogawa *et al.*: "Dense Fuel Cycles for Actinide Burning and Thermodynamic Data base", Int. Conf. on Evaluation of Emerging Nuclear Fuel Cycle Systems," GLOBAL'95", Vol. 1, p. 207 (Versailles, 1995).
- (11) Y. Arai, Y. Suzuki, and M. Handa: "Research on actinide mononitride fuel", Int. Conf. on Evaluation of Emerging Nuclear Fuel Cycle Systems," GLOBAL'95", Vol. 1, p. 538 (Versailles, 1995).
- (12) M. Mizumoto *et al.*: "Development of a High Intensity Proton Accelerator", Proc. 2nd

International Conference on Accelerator Driven Transmutation Technologies and Applications (Kalmar, 1996).

- (13) J. Kusano, *et al.*: "Status of the high intensity proton linear accelerator development in JAERI", *paper to be presented at this meeting* (1996).
- (14) T. Nakagawa *et al.*: "Nuclear Data for OMEGA Project", Proc. 3rd OECD/NEA Int. Information Exchange Mtg. on P-T, p. 371 (Cadarache, 1994).
- (15) T. Mukaiyama *et al.*: "Actinide integral measurements in FCA assemblies", Proc. Int. Conf. Nuclear Data for Basic and Applied Science, Vol. 1, p. 483 (Santa Fe, 1985),
- (16) H. Takada, *et al.*: "Production of Radioactive Nuclides in a Lead Assembly with 500 MeV Protons", Proc. Specialist's Mtg. on Accelerator-Based Transmutation" (PSI, 1992).

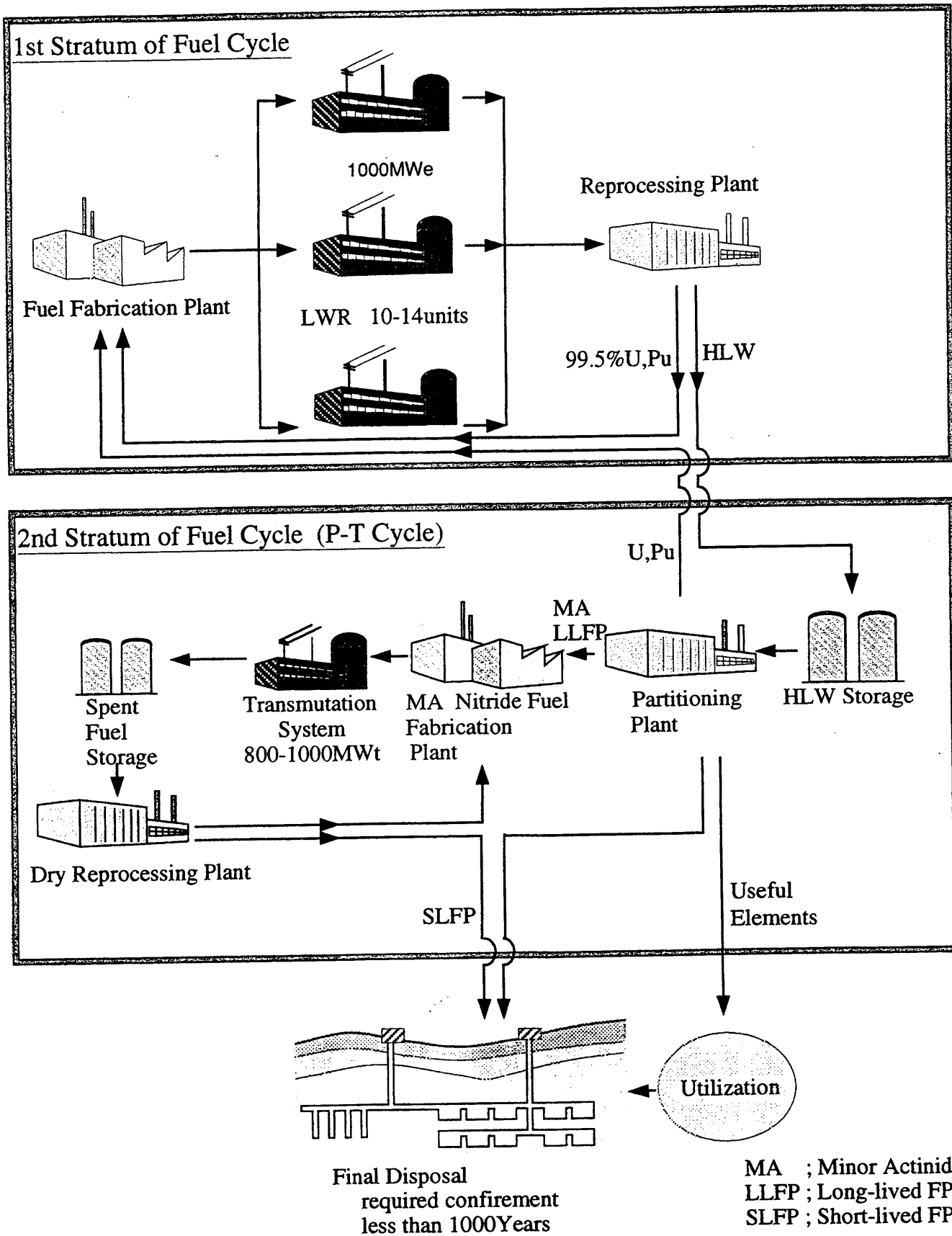


Fig. 1 JAERI's Concept of Double Strata Fuel Cycle for Complete HLW Management

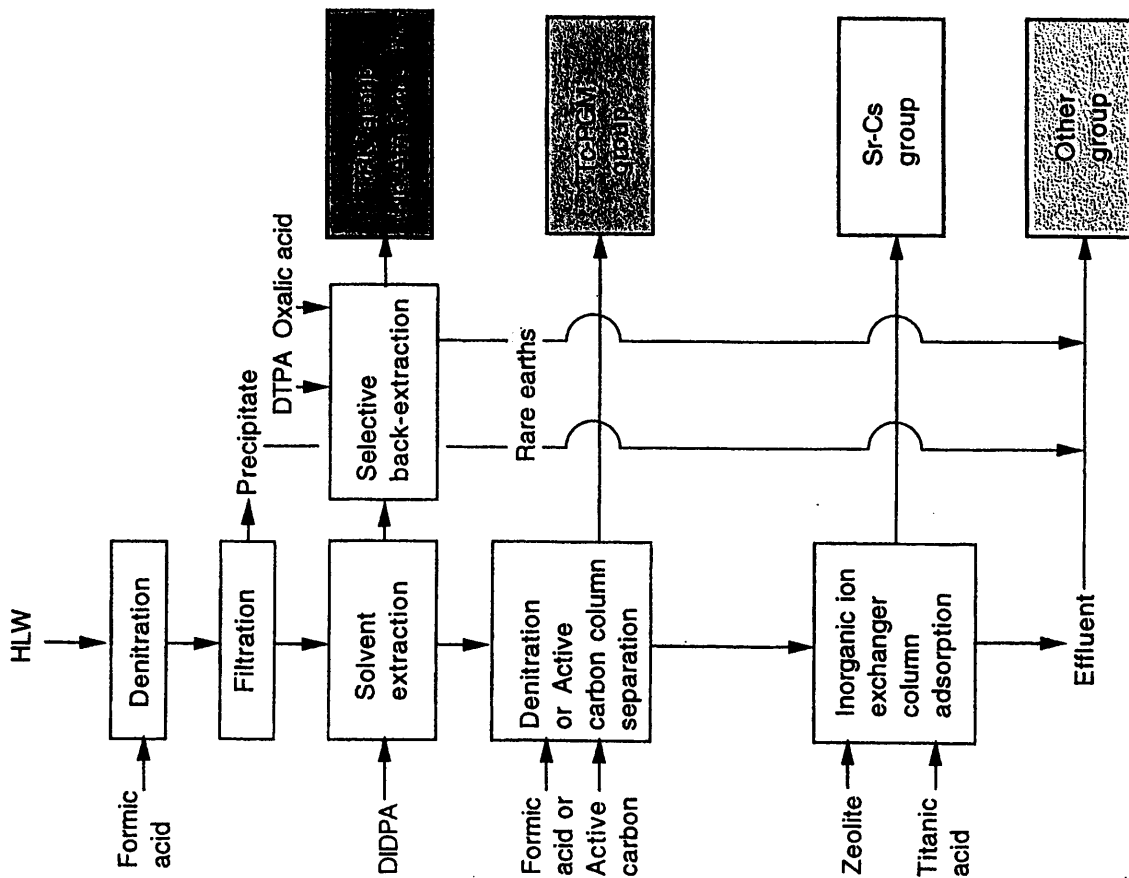


Fig. 2 Flow Sheet of Four Group Partitioning Process

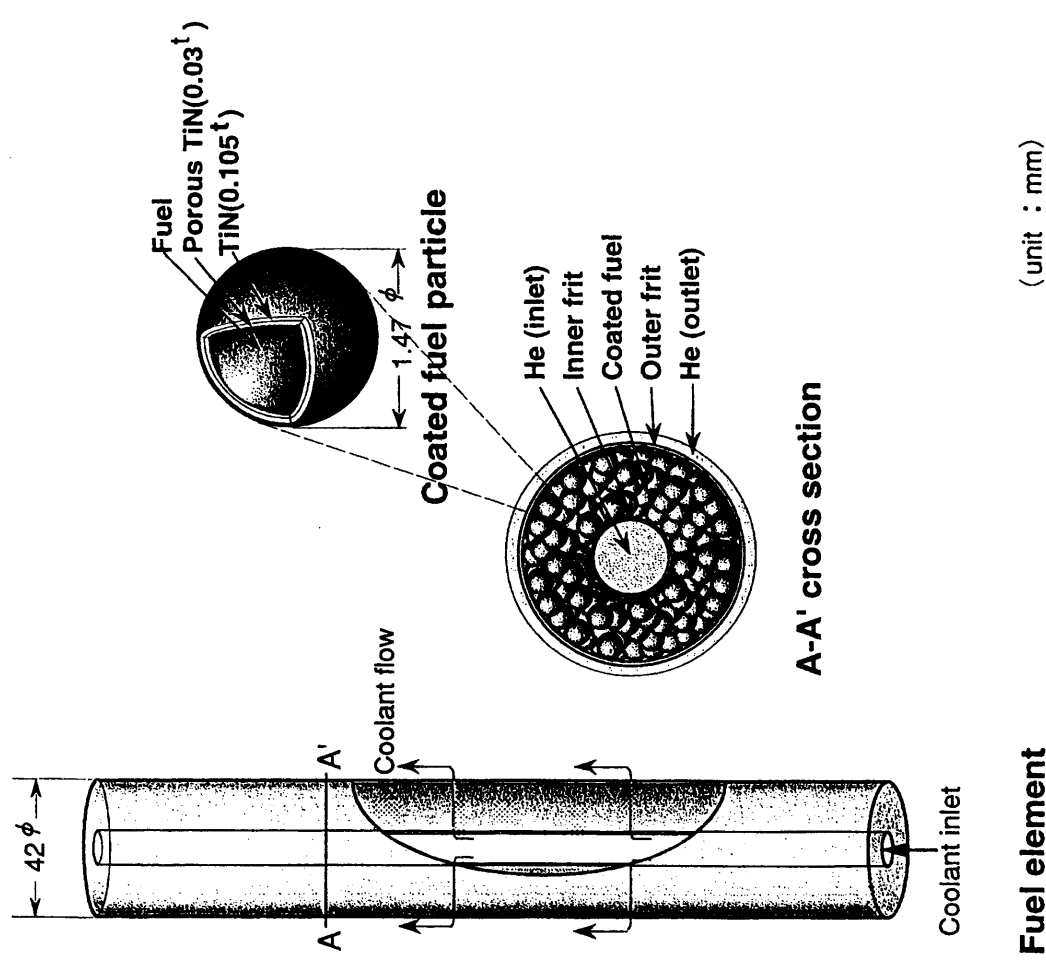


Fig. 3 Fuel element and coated fuel particle for He gas cooled reactor

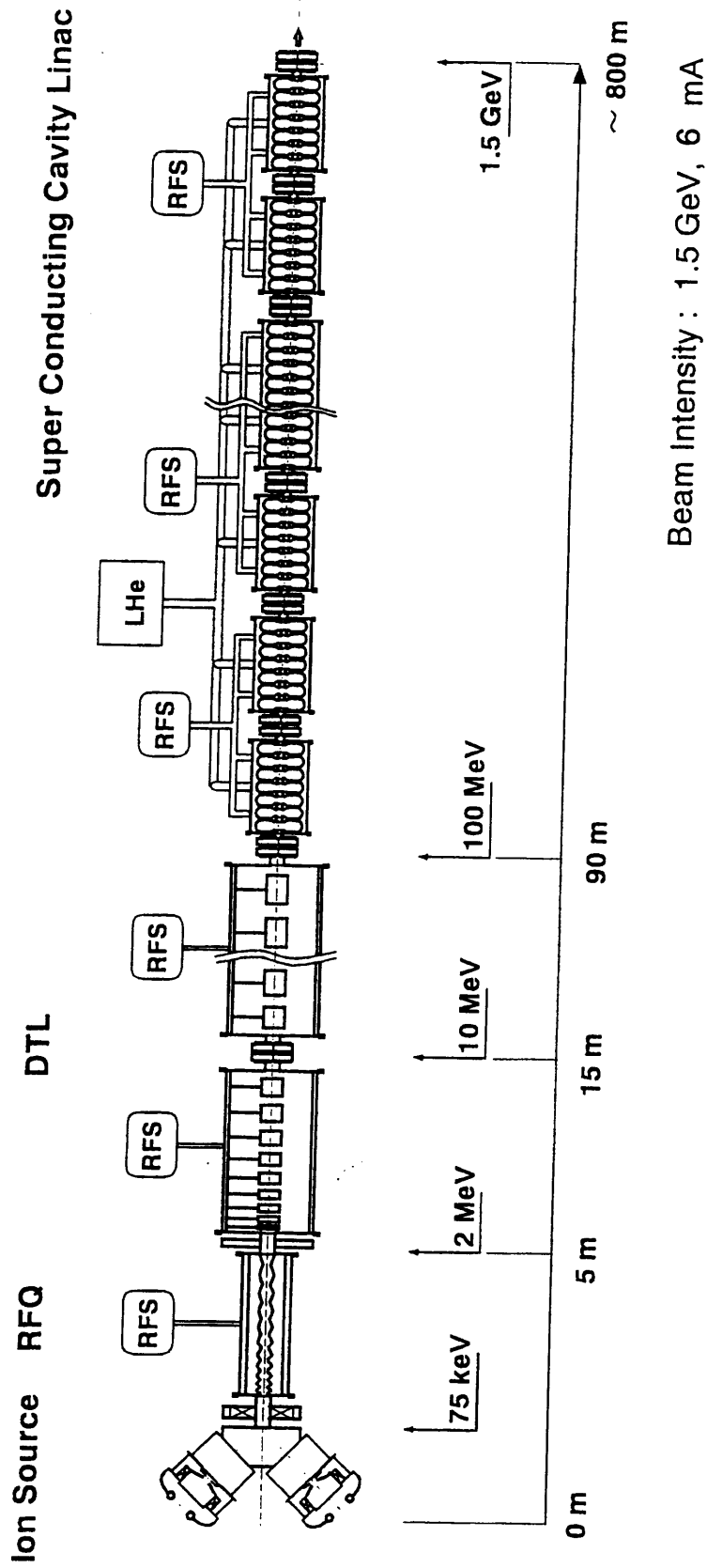


Fig. 4 Conceptual Layout of Intense Proton Accelerator with Superconductive Cavity

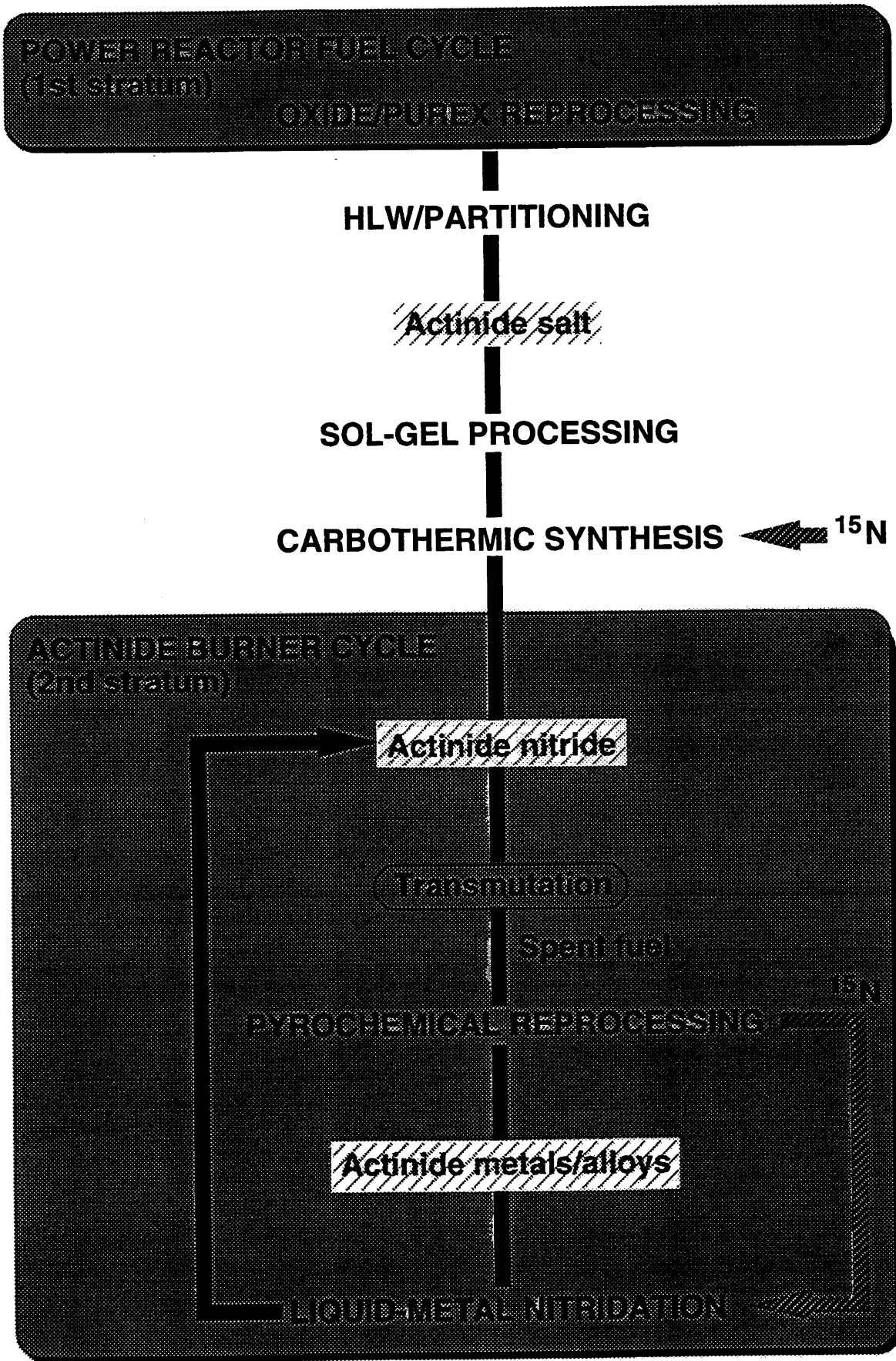


Fig. 5 Double Strata Fuel Cycle with Nitride/Pyrochemical Process

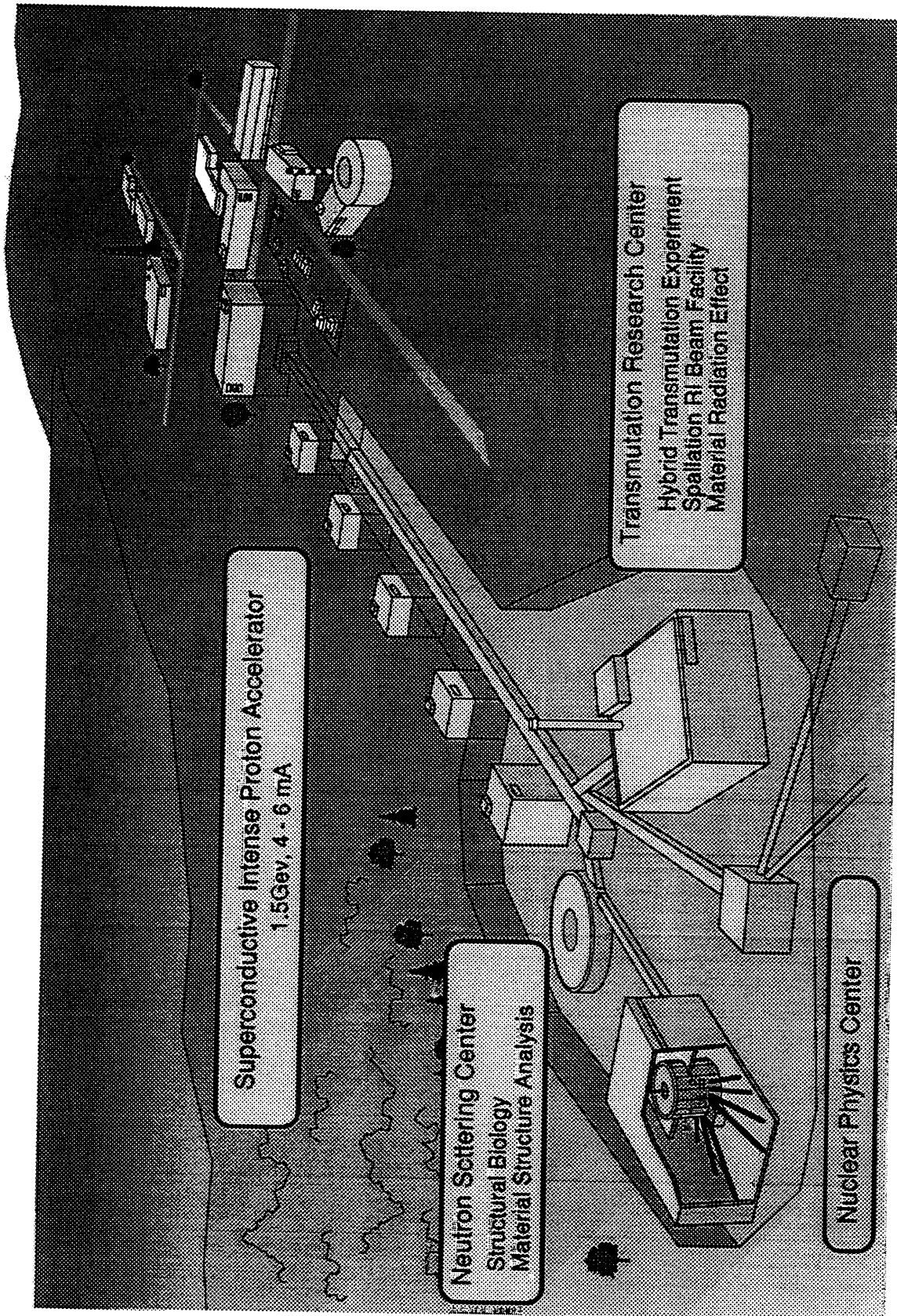
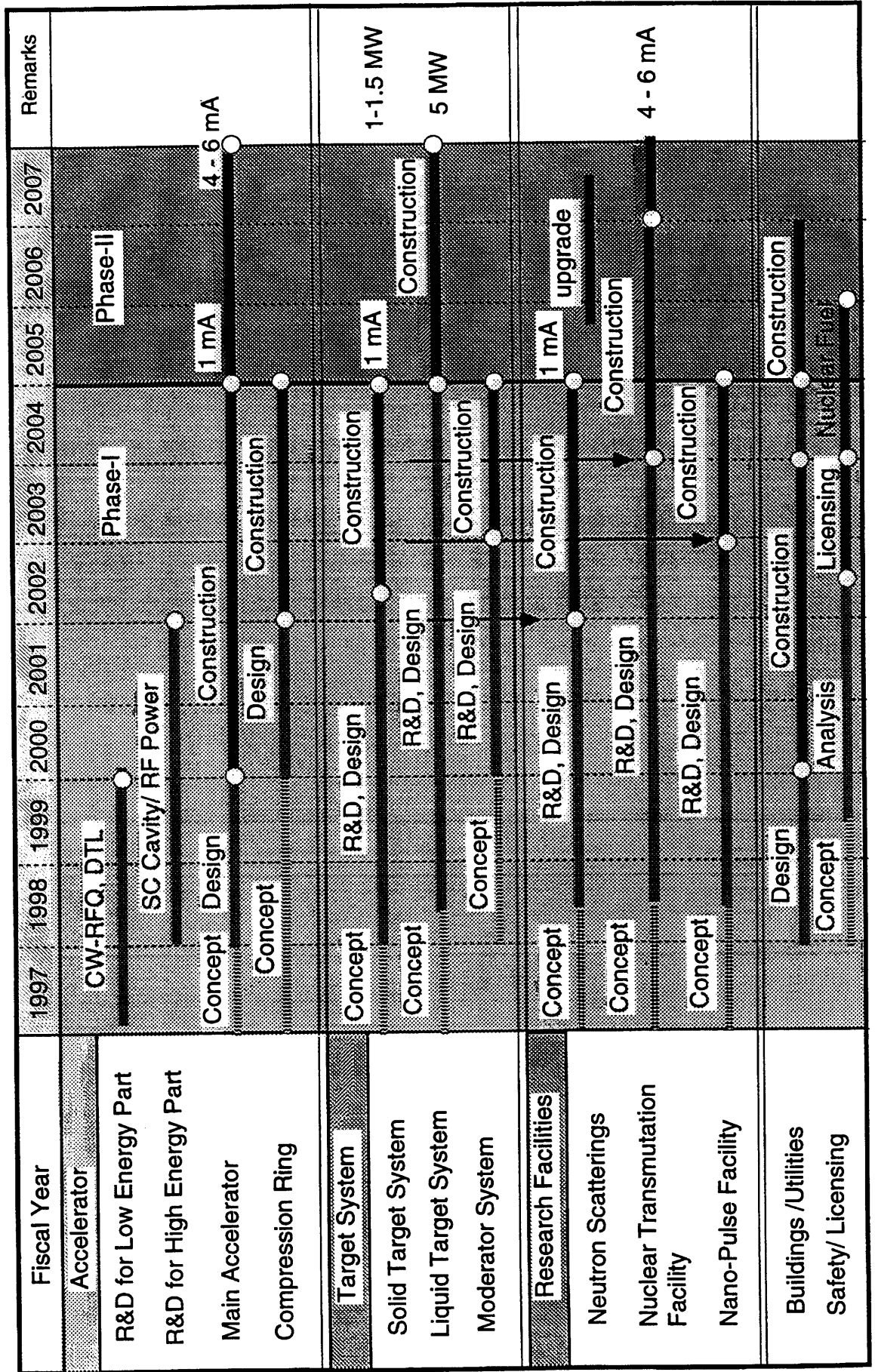


Fig. 6 Layout of proposed Facilities for Neutron Science

Fig. 7 Proposed Schedule for Construction of Neutron Science Research Facilities



PARTITIONING AND TRANSMUTATION OF NUCLEAR WASTE: THE DUTCH RAS PROGRAMME AND ITS RELATION TO INTERNATIONAL STUDIES

R.J.M. Konings, H. Gruppelaar, E.H.P. Cordfunke and J. Prij
*Netherlands Energy Research Foundation ECN,
P.O. Box 1, 1755 ZG Petten, Netherlands*

ABSTRACT

In the present paper a review of the Dutch RAS programme is presented. The objectives and achievements of the programme are described. Special attention is given to a recent assessment of the international status of P&T that has been made for the Dutch authorities. The major conclusion of this assessment is that P&T is an important instrument in the management of nuclear waste to achieve the technical limits (ALARA) with respect to radiotoxicity of the waste inventory as well as radiological effects of the disposal. The technology for P&T is, however, only partially available at present and recommendations for the directions of the RAS programme are given.

1. INTRODUCTION

Spent fuel elements from nuclear power plants contain many radioactive elements (actinides, fission products) which are a possible threat to mankind if released to the biosphere. Disposal in geological repositories is considered to be the final destination of spent fuel or the waste produced by reprocessing of the fuel elements, but is, however, not considered safe by a significant fraction of the population in many countries. This could be overcome if the actinides can be removed efficiently from the spent fuel and the radiotoxicity of the waste can be reduced to that of the uranium ore it was produced from, in a reasonable period of time (e.g. 250 years). Reprocessing, partitioning, recycling and transmutation are techniques to achieve this goal.

In 1991 the Netherlands Energy Research Foundation ECN started a research programme on recycling and transmutation of long-lived nuclides that are present in the spent fuel from nuclear power generation. This programme, which is known under the Dutch acronym RAS, is concentrated on the following five topics:

- reactor physics and scenario studies,
- chemical and material studies of fuels and targets,
- irradiations in the High Flux Reactor at Petten,
- radiological effects and risks,
- non-proliferation.

In the present paper a short description of the objectives and achievements of the RAS programme is given, followed by the present views on the state-of-the-art of partitioning and transmutation of actinides and fission products. Strategies and (innovative) fuel cycle technology required for the recycling of plutonium, minor actinides and fission products are discussed and their possibilities and limits are identified. Also the potential of future options with low actinide production (thorium cycle, accelerators) is considered. An extended report on this work is given in Ref. [1].

2. OBJECTIVES AND ACHIEVEMENTS OF THE RAS PROJECT

In the Dutch RAS programme the ALARA principle is applied to the radiotoxicity of the waste as well as to the risk of disposal to the benefit of future generations. As a consequence, the investigations in the field of P&T are focused on:

1. the actinides, in particular Pu and Am, which dominate the medium- and long-term radiotoxicity of HLW,
2. long-lived mobile components (in particular Np, Tc, I, Cs), which dominate the long-term radiological effects of disposal.

However, the justification of the work is not based on scientific arguments since the risks of geologically stored waste to future generations are estimated to be very low. The RAS programme should rather demonstrate the maximum possible measures that are necessary to relief public concern.

In the first phase of the RAS programme considerable attention was given to the transmutation of the fission products technetium and iodine. In the frame of the EFTTRA cooperation [2], an irradiation experiment (EFTTRA-T1) was performed in the HFR to study the behaviour of several target materials [3-5]. In parallel, scenario studies for the transmutation of these fission products in various reactor types have been made [6,7]. The overall conclusion of the work is that transmutation of technetium is technically possible although the irradiation times in most reactor types are (very) long and the technetium inventories are high. The transmutation of iodine in existing fission reactors does not look feasible (see below).

Currently, the RAS programme is focused on the transmutation of actinides. Different recycling modes for plutonium and americium are being studied by calculational methods and experiments, including MOX fuel in current LWRs and TRU fuels in future reactors. Again, the efforts within the EFTTRA programme are an essential part of this work: three irradiation experiments are planned in the HFR (Table 1) to study the irradiation behaviour of various ceramic materials that can be used as inert matrix in non-fertile fuels or targets [8]. In addition, laboratory experiments are performed to characterize a number of inert-matrix materials and to determine their physico-chemical properties. Various scenarios for the transmutation of plutonium and americium have been analysed [9-11]. An overall conclusion is not yet available but the preliminary results show that recycling of Pu is possible, whereas recycling of Am is only feasible if the technological problems with respect to reprocessing and fuel fabrication can be solved, as will be discussed below.

The use of the thorium cycle in fission reactors as a means to minimize the production of actinides has also been investigated. The state-of-the-art of the technology of thorium-based fuel has been reviewed [12] with respect to reactor physics, chemical and material aspects and actinide waste. In this study it was concluded that a substantial reduction of actinide radiotoxicity can be achieved by using thorium-based fuels if efficient partitioning and multiple recycling of uranium and thorium can be realised. Finally, a limited effort is devoted to the study of accelerator-based transmutation, including contributions to nuclear data for these systems [13].

In 1996, an assessment of the international status of P&T has been made for the Dutch authorities in order to define the next phase of the RAS programme [1]. The report has been reviewed by a national committee of experts from government, science and industry. In the following three sections, the views expressed in this report are summarized.

Table 1. Irradiation programme in the HFR at Petten.

Experiment	Description	Period
EFTTRA-T1	transmutation of the fission products technetium (~6% burn-up) and iodine	1994
EFTTRA-T2	<ul style="list-style-type: none"> • re-irradiation of technetium (> 20% burn-up) • irradiation of ceramic materials for the development of fuels for actinide burning (inert matrices) 	1996-1998
EFTTRA-T3	irradiation of inert matrices, mixed with enriched uranium	1996-1997
EFTTRA-T4	demonstration of the transmutation of americium in an inert matrix	1996-1999

3. STATUS OF PARTITIONING AND TRANSMUTATION

3.1 Plutonium and Uranium

Separation of plutonium (Pu) and uranium (U) from spent fuel will lead to a decrease of the radiotoxicity of the remaining waste by a factor 10. Using current PUREX technology, Pu and U can be separated from spent fuel with high efficiency (99.5-99.88 %). From the point of view of P&T, further improvement of the separation efficiency is only useful if also the minor actinides, in particular americium, are removed from the high level waste.

Pu and U can be recycled in thermal reactors in the form of mixed oxide (MOX) fuel. In case of 30 % MOX loading, as is current practice, Pu recycling in LWRs will slow down the growth of plutonium stocks. Higher MOX loadings (up to 100 %) will lead to net Pu consumption in future reactor designs. The effect of Pu recycling in LWRs on the radiotoxicity is, however, limited due to build-up of non-fissile Pu isotopes. Ultimately, the use of reactors/devices with fast neutron spectra is inevitable to reach substantial reduction of the plutonium radiotoxicity. The technology for the design of devices and fuels with maximum incineration rates needs to be developed.

3.2 Minor Actinides

Partitioning and transmutation of minor actinides, and especially americium, is necessary because a reduction of the radiotoxicity greater than 10 is wanted. If separation is realised with a 99-99.9% efficiency, a reduction of the radiotoxicity by a factor greater than 100 is within reach, provided that recycling is performed efficient as well. However, at present partitioning of the minor actinides americium and curium from PUREX waste solutions is not yet possible at an industrial scale. In several countries liquid-liquid extraction processes have been developed (Table 2) but in all cases the lanthanides, an important group of fission products, are co-extracted as a result of their chemical similarity to the trivalent actinides. New techniques to separate the trivalent actinides and lanthanides as well as the actinides mutually are required. The use of macrocyclic extraction molecules in Supported Liquid Membranes (SLM) is a promising development in this field and is studied in several European laboratories, among which ECN.

Transmutation of the minor actinides can be done efficiently in a fast-neutron flux but also the benefits of a "once-through" incineration in a thermal flux have to be evaluated. Neptunium can be recycled in MINOX fuels, americium in special targets containing an inert matrix. In practice, recycling of curium does not seem feasible at the moment because of the α -, γ - and neutron doses due to decay and spontaneous fission which extremely complicate the fabrication curium fuels and targets.

When transmutation of the minor actinides is introduced in the fuel cycle, existing non-proliferation measures have to be extended to cover P&T. Most of the problems to the minor actinides are similar to those of reprocessing and MOX fabrication plants which have been proven to be adequately safeguardable. However,

Table 2. Liquid-liquid extraction processes for the trivalent actinides.

process name	country	extraction molecule	HNO ₃ concentration	typical results
DIAMEX	France	diamides	0.5-5 M	Am+Cm > 95%
DIDPA	Japan	DIDPA/TBP	0.5 M	Am+Cm > 99.9%
TRPO	China	TRPO	0.1 - 1 M	Am+Cm > 95%
TRUEX	USA	CMPO/TBP	0.1 - 3 M	Am+Cm > 99.9%

specific P&T issues need to be addressed, such as the potential of mis-use of minor actinides and the new facilities. Until today the civil fuel cycle has shown to be a rather difficult route for proliferation and it is of utmost importance to maintain this characteristic also for new installations and fuel cycles.

3.3 Fission Products

Partitioning and transmutation of long-lived fission products is only relevant from the point of view of reduction of radiological effects. In this respect, the following three fission products need to be considered: cesium (^{135}Cs), iodine (^{129}I) and technetium (^{99}Tc). The technology to separate these elements from HLLW is available on a laboratory scale, but has not been implemented in the PUREX process. Technetium and iodine can be transmuted by single neutron capture, but the transmutation half-lives and inventories in most reactor types are high. In addition, transmutation of these fission products requires additional enrichment. Transmutation of cesium is not feasible because of the low neutron absorption cross section of ^{135}Cs and parasitic absorptions in ^{137}Cs and ^{133}Cs which are present in the fuel in about equal quantities as ^{135}Cs .

3.4 Survey of transmutation devices

Recycling of plutonium is possible in existing LWRs with respect to reduction of masses. However, as multiple recycling of Pu in LWRs is limited and transmutation of minor actinides in existing thermal reactors is not effective, a significant radiotoxicity reduction is only obtained in follow-up transmutation in dedicated burners. These burners should have low Pu production, a high specific power and for minor actinide incineration, a fast neutron spectrum. Nearby technology indicates CANDU and CAPRA burner types as interesting candidates; future developments hint in the direction of accelerator-based systems. Also fission-product transmutation can best be achieved in dedicated thermal burners or in moderated subassemblies of fast reactors since transmutation half-lives and inventories in LWRs are very high. The potential of existing, near-by and future reactor technology is summarized in Table 3.

Table 3. Review of the recycling capabilities of various transmutation devices; MA denotes minor actinides, FP fission products.

device	Pu	MA	FP
LWR	limited ^a	poor	no
HWR	moderate ^{a,b}	poor	moderate
HTR	limited ^c	poor	no
ALMR	good	good	no
CAPRA	excellent	good	reasonable ^d
Pb-cooled burner	excellent	good	reasonable ^d
molten salt reactor	good?	?	?
accelerator-driven thermal burner	good?	?	good
accelerator-driven fast burner	excellent	good	?

^a Multirecycling of MOX is limited due to production of even-mass Pu and other actinides.

^b 100% Pu in an inert matrix.

^c Reprocessing is difficult.

^d In a moderated subassembly.

4. OPTIONS WITH LOW ACTINIDE PRODUCTION

4.1 Radiological clean energy production?

The use of pure fissile material, in particular fissile U isotopes, gives lowest long-lived radiotoxic actinide production. This requires fissile material from either (high) enrichment or breeding. Use of uranium, highly enriched in ^{235}U (HEU), is not recommended from the point of view of non-proliferation. The alternative is the use of ^{233}U produced in the thorium cycle, which is more proliferation resistant, due to hard γ -rays from co-generated ^{232}U . At present, extension of burnup of LWR fuel is a good measure to limit the actinide production per unit of generated electricity in the current open U/Pu cycle.

4.2 Thorium cycle in reactors

The effective use of thorium in an open cycle requires fissile topping material and a high burnup. The radiotoxicity of waste is lowest if ^{233}U or ^{235}U (HEU) is used as topping material. Further reduction of the radiotoxicity is obtained in a closed cycle using thermal reactors and ^{233}U or ^{235}U (HEU) as topping material and recycling of uranium. The best result should be obtained in a fast reactor with recycling of all actinides and without topping material. The main obstacle for introduction of the closed cycle is that reprocessing and fuel fabrication have to be introduced on an industrial scale, including adequate safeguards. A good introduction to the thorium cycle could be to burn Pu in a thorium oxide fuel matrix.

4.3 Accelerator-based energy production

Accelerator-based systems have probably some safety advantages and produce excess neutrons. These two aspects give flexibility to design future systems for safe, clean and acceptable nuclear energy production and/or waste transmutation. The accelerator offers the possibility for applying a closed thorium cycle, due to excess neutrons (when coupled to thermal reactors) or additional safety (when coupled to fast reactors). The Fast Energy Amplifier is one of the examples with high potential. Other advanced features are related to waste transmutation of difficult long-lived components, like minor actinides and long-lived fission products and the possibility of incinerating the radioactive inventory at the end of a nuclear era.

5. CONSEQUENCES FOR DISPOSAL

The radiological effects of disposal of waste in a (geological) repository are not only determined by the radiotoxicity but also by the performance of the natural and engineered barriers of the disposal concept. The safety-relevant radionuclides that are identified 'normal evolution' release scenarios used in performance assessments, are the fission products ^{135}Cs , ^{129}I , ^{79}Se , ^{99}Tc and the actinide ^{237}Np and its decay products. However, the calculated maximum exposures in 'altered evolution' scenarios are far below the natural background. For human intrusion scenarios the maximum exposure is more significant and in these scenarios the actinides ^{241}Am , ^{243}Am and ^{240}Pu predominate the radiological effects.

Table 4. Some results of performance assessments for disposal of vitrified HLW in a salt formation in the Netherlands.

Scenario	Maximum exposure		Dominant radionuclides
	magnitude (Sv/y)	time (y)	
Normal evolution	10^{-7}	10^6	^{237}Np , ^{233}U , ^{135}Cs
Ground water intrusion	10^{-8}	10^6	^{135}Cs , ^{79}Se , ^{226}Ra
Human intrusion	10^{-4}	500	^{241}Am , ^{243}Am , ^{240}Pu

It should be realised that the separation of the actinides alone, as a result of which the radiotoxicity of the waste is reduced below the level of natural uranium ore after 250 years, does not imply that disposal in a (geological) repository is no longer needed. But if such waste is disposed in a carefully selected geological formation, the risk can be as low as that of natural uranium ore in its geological situation.

6 CONCLUSIONS

Partitioning and transmutation of actinides are important elements in the management of the waste from nuclear power generation. They are means to achieve the limits (ALARA) with respect to the radiotoxicity of the inventory of disposal sites as well as the radiological effects of human intrusions of disposal sites in the future. The technology required for the implementation of P&T of actinides in the fuel cycle is only partially available at present and much research is still needed. In the coming years the efforts of the RAS programme should be concentrated on:

- Improved partitioning methods for trivalent actinides.
- Transmutation of actinides using non-fertile fuels.
- Scenario studies using 100 % MOX, HWRs and fast burners.
- Innovative systems for future "clean" energy production and transmutation using the thorium cycle and/or accelerators.

To reduce the (small) effects of exposure of radionuclides due to normal evolution, transmutation of long-lived fission products should also be realised. However, on the basis of the present technology this does not seem feasible and partitioning and immobilisation might be a more realistic way of achieving this. Study of this topic is recommended.

ACKNOWLEDGEMENT

Dr. J.L. Kloosterman is acknowledged for his critical comments and his willingness to present this paper at the meeting.

REFERENCES

1. H. Gruppelaar, R.J.M. Konings, E.H.P. Cordfunke, J. Prij, *Transmutation of nuclear waste: State-of-the-art of the international research and strategy studies on partitioning and transmutation of actinides and fission products*. Report ECN-R--96-006 (1996).
2. J.-F. Babelot, H. Gruppelaar, G. Mühling, C. Prunier, M. Rome, M. Salvatores, *EFTTRA, a European collaboration for the development of fuels and targets for transmutation*. Proceedings of the Third International Information Exchange Meeting on Actinide and Fission Product partitioning and Transmutation, Cadarache, December 12-14, 1994, NEA/P&T Report N/13, 1995, pp.110-115.
3. R.J.M. Konings, E.H.P. Cordfunke, G. Dassel, W.M.P. Franken, H. Gruppelaar, J.L. Kloosterman, R. Conrad, *Irradiation tests of transmutation of the fission products iodine and technetium*. Proceedings "Global 1995 - International Conference on Evaluation of Emerging Fuel Cycle Systems", Versailles, September 1995, p. 1631-1637.
4. R.J.M. Konings, W.M.P. Franken, R. Conrad, J.-F. Guegnon, J.-C. Spirlet, *Transmutation of technetium and iodine - Irradiation tests in the frame of the EFTTRA cooperation*. Nucl. Technol., submitted.
5. R.J.M. Konings, *Transmutation of iodine: results of the EFTTRA-T1 irradiation test*. J. Nucl. Mat., submitted.
6. J.L. Kloosterman, J.M. Li, *Transmutation of Tc-99 in fission reactors*. Proceedings of the Third International Information Exchange Meeting on Actinide and Fission Product partitioning and Transmutation, Cadarache, December 12-14, 1994, NEA/P&T Report N/13, 1995, pp.285-297 (also Report ECN-RX--94-119).
7. J.L. Kloosterman, J.M. Li, *Transmutation of Tc-99 and I-129 in fission reactors - A computational study*. Report ECN-RX--95-002 (1995).
8. J.-F. Babelot, R. Conrad, H. Gruppelaar, G. Mühling, C. Prunier, M. Salvatores, G. Vambenepe, *EFTTRA, irradiation experiments for the development of fuels and targets for the transmutation*.

- This meeting.
9. J.H. Bultman, *Actinide transmutation in nuclear reactor systems*. PhD Thesis, Delft University of Technology, (1995).
 10. J.L. Kloosterman, J.M. Li, *Transmutation of americium in fission reactors*. Proceedings "Global 1995 - International Conference on Evaluation of Emerging Fuel Cycle Systems", Versailles, September 1995, p. 1106-1113.
 11. J.L. Kloosterman, W.J.M. de Kruijf, *Transmutation of americium in thermal reactors*. Proceedings of the Third International Information Exchange Meeting on Actinide and Fission Product partitioning and Transmutation, Cadarache, December 12-14, 1994, NEA/P&T Report N/13, 1995, pp.276-284 (also Report ECN-RX--94-116).
 12. W.M.P. Franken, J.H. Bultman, R.J.M. Konings, V.A. Wichers, *Evaluation of thorium based nuclear fuel - Extended summary*. Report ECN-R--95-006 (1995).
 13. A.J. Koning, H. Gruppelaar, *Nuclear data for hybrid nuclear systems*. Proceedings "Global 1995 - International Conference on Evaluation of Emerging Fuel Cycle Systems", Versailles, September 1995, p. 855-862.

**TRANSMUTATION OF TRANSURANICS: NEUTRONICS, ACTINIDES BALANCE,
SAFETY AND FUEL PROVISION ASPECTS**

N.S.Robotnov

State Research Center -A.I.Leipunskiy Institute of Physics and Power Engineering,
249020, Bondarenko Sq.1, Obninsk, Kaluga Region, Russia

Abstract

A review is given of the selected works on the subject in Russia with the emphasis on the latest results published or presented after GLOBAL95 Conference. The main attention is concentrated on following problems: evolution of the actinides isotopic mixtures during multiple recycling in thermal and fast neutron systems operating separately or in combinations; compatibility of the transmutation with the breeding of secondary fuel; influence of plutonium composition, including weapon-grade, on the recycling process; nuclear safety related problems of actinides recycling; nuclear data for transmutation; criteria of assessment of radiological and environmental consequences of actinides transmutation; excess neutrons available for transmutation in accelerator-driven facilities; activation of neutron producing accelerator targets.

Introduction

During last three years some new attitudes toward nuclear fuel cycle were developing fast as the new series of GLOBAL-93 and GLOBAL-95 [1,2] conferences clearly demonstrated. These new trends included: increased attention to transmutation of minor actinides and possibly of some fission products; fast disposition of the excess separated plutonium, especially weapon grade; extensive analyses of the transmutation and plutonium utilization potential of accelerator-driven facilities. These problems are closely interconnected and in Russia the interest in P&T research is mainly stimulated by the importance of plutonium problem.

In 1995 the IAEA has undertaken efforts to produce Status Report on Transmutation in Non-OECD Countries as an IAEA Technical Document. The text was compiled, corrected and approved last September and now is about to be published. It contains rather comprehensive general review of the progress in P&T covering the activity of about dozen largest Russian institutions during last few years. So present paper is focused on the results obtained quite recently and best known to the author which means mainly A.I. Leipunskiy IPPE publications. At first short general review is given and then a few points are outlined in more details.

The strategy of P&T research in Russia is determined by following considerations. Due to obvious slowing down of the development of nuclear power industry the balance of research efforts between evolutionary and innovative approaches shifted to the latter. In the domain of basically evolutionary designs, which means critical fission reactors with solid fuel and liquid coolant, an ideal facility included in closed fuel cycle with multiple recycling, aimed at as complete utilization of natural resources as possible, should have:

- all principal reactivity coefficients of proper signs and values;
- fuel burn-up not less than 100 GW days/thm;
- breeding ratio not less than 1.3;
- ability to burn, in equilibrium regime, all Np and Am of its own and, if needed, some quantity of MA from outside;
- proliferation resistant fuel cycle;
- transparent ability to withstand design basis accidents without catastrophic consequences;
- good economy.

If these requirements are conflicting or some of them just may not be realized then we should look into a broader domain of designs.

1. General review

Transmutation related research at IPPE connected with the topic of present paper consists of following interacting sectors:

- analysis of the status and prospects of closed fuel cycles technologies aimed at transmutation of minor actinides and probably selected fission products;
- extensive calculations of the properties and parameters of the plutonium fueled fast reactor cores used as actinide burners;
- experiments on critical facilities, imitating the cores of large fast reactors, coordinated with the calculations and analysis programs and used to verify both input data and calculation methods.
- measurements and evaluation of basic microscopic nuclear data;
- designing, related experiments and calculations on the neutron producing accelerator targets of accelerator-driven transmutation facilities.

Present paper deals mainly with the reactor side of the transmutation problem because in ADTT research in Russia the Institute of Theoretical and Experimental Physics (Moscow) is main driving and coordinating force as demonstrated by the latest ADTT Conference in Kalmar [3]. But some important work is done in IPPE also and those results are discussed briefly.

1.1 Closed nuclear fuel cycle as the way of developing long term sustainable nuclear power industry.

Mining of uranium does not satisfy present demand and situation will get worse in coming 15 years with fuel fabrication relying significantly on the stored uranium reserves. Spent fuel reprocessing rate is only a fraction of the discharge rate and does not prevent piling up of large masses of SNF consisting of almost untouched natural uranium with uncertain future. So a time will come, most probably in the second half of the next century, when nuclear power industry will face following situation in raw materials: millions of tons of depleted uranium and a limited quantity of assorted plutonium and minor actinides, mostly in the spent fuel

form. Let's call the time "Day X". Sustainable level of nuclear power after the Day X, $P(t > X)$, depends on the quantity $M_{Pu}(X)$ and quality of plutonium available determined by the global scenario of plutonium utilization which is combination of national scenarios, formed today and varying drastically. $M_{Pu}(X)=0$ means $P(t > X)=0$ i.e. the end of the nuclear power industry based on critical fission reactors because no system critical on depleted uranium only is possible. We made an attempt to estimate X and $P(X)$ for Russian national nuclear power industry and to investigate the sensitivity of these values to some features and parameters of the selected scheme of the nuclear fuel cycle within different concrete scenarios [4]. Basic assumptions are:

- thermal and fast reactors are used in varying proportion;
- regenerated uranium is recycled both in thermal and fast reactors;
- plutonium is recycled in MOX fuel of fast reactors;
- the possibility of utilization of excess weapon-grade plutonium is considered.

The results indicate that:

- in the model adopted X is close to 2080;
- $P(X)$ is considerably lower than the maximum value of mainly uranium nuclear power capacity reached earlier;
- weapon-grade plutonium is essential; if added to reactor-grade plutonium as 20 per cent admixture and used in fast reactors with breeding ratio 1.3 it may add up to 30% to $P(X)$;
- BR=1.3 increases $P(X)$ by some 70 per cent as compared to the case with BR=1.0.

Delay of the introduction of fast breeders into national nuclear power industry obviously decreases $P(X)$ considerably. From the point of view of nuclear power industry development in the next century there is no plutonium surplus in Russia. The same well may be true in global scale if present trend to develop primarily net plutonium burners prevails (warnings of this kind may be found elsewhere, see, for example, [5]). The best way to deal with weapon-grade plutonium is to convert it to "spent fuel standard" in fast breeders with as good breeding ratio as possible and keep the spent nuclear fuel for later recycling, thus solving non-proliferation problem and using plutonium's energy producing potential.

Pu recycling and breeding may be combined with MA burning neutronically but safety should be studied further; already nearest generation of new reactors with life-time expectancy of at least 50 years should be designed flexible to meet the requirements of steadily growing plutonium breeding when necessary. Recycling of U in thermal reactors increases total electricity production during the considered period by 14 per cent, recycling of Pu in thermal reactors - by further 4 per cent only. Introduction of fast reactors with BR=1.0 brings 21 per cent increase which rises to 43 per cent at BR=1.3.

1.2 Recycling of plutonium and minor actinides: nuclear safety related consequences.

Plutonium and minor actinides may be recycled in both thermal and fast reactors. The efficiencies of these two options depend not only on the physics and technology of corresponding systems but on many varying and not always predictable external conditions like availability of competing energy sources, natural uranium prices, public acceptance, general national energy policies etc. But there is a set of much more definite safety related problems which should be considered first:

Core neutronics: worsening of Doppler and void reactivity effects, control rod worth, delayed neutrons parameters.

Power density distribution: sharper non-uniformities starting at the pellet level; strong and non-monotonous time-dependence; possible complications of loading and reloading schemes.

Accidents development: higher recriticality dangers in a case of melt down with high Pu content; worse kinetic parameters with resulting increase of the energy release during an excursion.

Proliferation issues: separated plutonium of practically any composition is most proliferation sensitive substance unlike plutonium in spent fuel.

All these aspects are closely investigated and some of the latest results are to be presented in a few papers at PHYSOR-96 Conference, September 16-20, 1996, Mito, Japan. [6-8].

Experience with liquid metal coolants in IPPE is not restricted by sodium cooled fast reactors but includes the reactors cooled by liquid heavy metal, lead-bismuth eutectics first of all. This concept is revisited nowadays in connection with the possibility to use fast reactors cooled by heavy metals for Pu utilization and MA transmutation (see, for example [9-10]).

1.3. Experiments on BFS facilities.

Actinide integral measurements were carried out on BFS similar to those on a set of FCA-9 assemblies to test the fission and capture cross sections of minor actinides (MA) described in [11]. The integral data measured are:

- the central fission rate ratio (FRR);
- the central sample worthies (CSR).

The core of BFS-67 assembly was composed with 96% enriched metal plutonium, depleted uranium dioxide, sodium and stainless steel. This composition was similar to the SUPERPHENIX core. About half of uranium dioxide in this composition was replaced with sodium for constructing of BFS-69 assembly core.

For both BFS assemblies the spectra were similar to the spectrum averaged over FCA assemblies. The integral data measured are:

- FRR of Np-237 and Pu-239 relative to fission in U-235 ;
- CSW of Np-237 ,Pu-239 and U-235.

All samples sizes were less than in FCA experiments. The Np and Pu samples were of three different size. The exact description of the assemblies, experimental devices and obtained results of measurements were given in [12]. Some of the results provided by the authors are given in Tables 1-4.

The starting point of neutron data testing is homogeneous calculation of FRR and first order perturbation theory using ABBN approach [13] . Evaluation of experiments means taking in account the heterogeneous structure of core cells, finite sizes of samples and group constant correction at calculation of CSW.

Heterogeneous structure of critical assembly's cell is taken into account by using the integral-transport approximation. Undisturbed group fluxes and adjoint fluxes are obtained from solutions of corresponding integral-transport equations in the cell approximation. Criticality is attained by modification of a neutron leakage. Perturbation of collision probabilities are taking in account too by calculation results using perturbation theory (first type of correction). Consideration of a detailed energy structure of adjoint solution gives the additional contribution into the reactivity worth ratio (second type of correction). Taking in account the finite size of samples have consisted the third type of correction.

The results of measurements and evaluation are presented in Tables 1 and 3 for CSW ratios of Pu - 239 / U - 235 , in Table 2 for CSW ratios of Np-237/Pu-239 and in Table 4 for CSW ratios of Np-237/U-235.

Table 1. CSW ratio U-235 / Pu - 239

Assemb. FCA	EXP. virgin	Correction of 1 and 2 types	Correction of 3 type for Pu	Correction of 3 type for U	EXP. evaluated
9-1	1.476	-0.023	-0.146	+0.109	+1.416
9-2	1.617	-0.010	-0.127	+0.124	+1.604
9-3	1.713	-0.009	-0.008	+0.016	+1.712
9-4	1.708	-0.006	-0.079	+0.089	+1.712
9-5	1.750	0	-0.075	+0.081	+1.756
9-7	1.745	-0.002	-0.065	+0.060	+1.738

Table 2. CSW ratio Np-237 / Pu - 239

Assemb. FCA	EXP. virgin	Correction of 1 and 2 types	Correction of 3 type for Pu	Correction of 3 type for U	EXP. evaluated
9-1	-0.865	+0.179	-0.220	-0.073	-0.979
9-1	-0.242	+0.072	-0.042	-0.013	-0.225
9-3	-0.014	+0.021	-0.014	+0.005	-0.002
9-4	+0.054	+0.006	-0.005	+0.006	+0.061
9-5	+0.158	+0.002	-0.004	+0.010	+0.166
9-7	+0.117	+0.001	-0.003	+0.007	+0.122

Table 3. CSW ratio Pu - 239/U - 235

Assembly	Type of sample	EXP VIRGIN	ZERO SIZE of samples	correction of 1 and 2 types	EXP evaluated
BFS 67	Pu - A	1.350 ± .020	1.327 ± .021		
	Pu - B	1.406 ± .008	1.365 ± .009		
	Pu - C	1.404 ± .005	1.352 ± .007		
	averaged value		1.353 ± .019	+ .011	1.364 ± .019
BFS 69	Pu - A	1.591 ± .018	1.560 ± .020		
	Pu - B	1.607 ± .007	1.552 ± .008		
	Pu - C	1.592 ± .004	1.528 ± .006		
	averaged value		1.541 ± .019	- .002	1.539 ± .019

Table 4. CSW ratio Np-237 / U-235

Assembly		VIRGIN EXP	ZERO SIZE of samples	correction of 1 and 2 types	evaluated experiment
67-1	Np - A	-.240 ± .011	-.250 ± .011		
	Np - B	-.228 ± .006	-.240 ± .008		
	Np - C	-.228 ± .005	-.245 ± .007		
	averaged value		-.245 ± .010	+ .023	-.222 ± .011
69-1	Np - A	-.120 ± .010	-.128 ± .010		
	Np - B	-.119 ± .006	-.130 ± .007		
	Np - C	-.114 ± .003	-.131 ± .004		
	averaged value		-.130 ± .005	+ .021	-.109 ± .006

1.4. Nuclear data measurements and evaluations.

Improvement of nuclear data for minor actinides is important for transmutation projects using actinide burner reactors and needed for the nuclides $^{237,238}\text{Np}$, $^{238,242}\text{Pu}$, $^{241,242,242\text{m},243}\text{Am}$ and $^{242,243,244,245,246}\text{Cm}$. Some of the most important cases are now investigated in the framework of ISTC Project-304 "Measurement and Analysis of Basic Nuclear Data for Minor Actinides". The latest works include:

- preliminary results of precise measurements of the fission cross sections of $^{244,245,246,247}\text{Cm}$ and $^{242\text{m}}\text{Am}$ by 0.15÷7.0 energy neutrons;
- first runs of measurements of secondary neutron spectra, fission product yields, delayed neutron yields and inelastic scattering cross section for Np-237;
- improved evaluations of the most important cross sections for Np-237.

Extensive calculations and evaluations of the nuclear data are now done for higher energy range to satisfy the needs of ADTT research and development. The latest results:

- special library is developed (MENDL-2, Medium Energy Nuclear Data Library) for the investigations of the activation and transmutation of the materials irradiated by the nucleons of intermediate energies (neutrons up to 100 MeV, protons up to 200 MeV, more than 100 000 reactions). First, neutronic, part is described in [14], proton part is to be converted to an agreed format and then released;
- systematics were developed of the cross sections of the threshold reactions on 14 MeV neutrons based on a new approach taking into account both equilibrium and non-equilibrium mechanisms of nuclear reactions [15-16].

1.5. Research on the accelerator targets for ADTT.

Efforts are undertaken in collaboration with Design Bureau "Gidropress" on the development of neutron producing liquid heavy metal targets irradiated by very powerful proton beams in GeV energy range. Both beam-window and windowless targets are investigated. This research includes:

- calculations of thermophysical and hydrodynamic properties of the targets and optimization of the designs;
 - material studies;
 - calculations of the diaphragm behavior, energy release, activation, including the accumulation of long-lived radionuclides, and gas production in liquid Pb-Bi and Pb targets;
 - release of radionuclides from liquid and solid targets.
- A few papers on these subjects were presented at Kalmar Conference [17-21].

2. Selected results on the actinides balance in various scenarios of Pu and MA recycling

2.1. Comparison of thermal and fast reactors in actinides recycling.

Transmutation of MA is closely linked with Pu utilization and would influence the actinide balance. The problem is whether MA burning is compatible with Pu breeding needed to support stable to say nothing of developing nuclear power industry. Another problem is whether nuclear safety requirements to Pu and MA containing cores may be met in fuel cycles involving Pu breeding. It's very wide domain so here we consider only some most prominent differences between thermal and fast reactors used for multiple long-term recycling of transuranics. A simple calculational model based on KARE code [22] was chosen to make the comparison (the results were presented in [4]). Basic features of the model are:

- the fuel of the first cycle is MOX-fuel, mixture of depleted uranium with reactor plutonium (isotopic composition of the latter is given in Table 5); enrichment by plutonium is sufficient to support chain reaction for planned campaign period and to reach planned burnup (parameters of the first cycle are given in Table 6);
- neutron spectra, fluxes and one-group constants coincide with those of BN-800 for fast reactor and with the parameters of Pu fueled VVER-1000 for thermal reactor;
- after each cycle the spent fuel is cooled for three years, fission products are eliminated completely, plutonium of initial composition and depleted uranium are added in the quantities necessary to complete the next cycle;
- all the actinides are left in refabricated fuel.

Table 5. Isotopic composition of fresh plutonium used in calculational model.

Isotope	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
Concentration, per cent	0.5	60.0	24.5	10.9	4.1

Table 6. Parameters of the first cycle.

Parameter of the cycle	Reactor type	
	Fast	Thermal
Time of irradiation, days	420	920
Pu enrichment, per cent	18.9	5.96
Neutron flux, $\text{cm}^{-2} \text{c}^{-1}$	$6.0 \cdot 10^{15}$	$4.0 \cdot 10^{14}$
Burn up, per cent of heavy atoms	6.6	3.6
Number of fissions/number of captures	0.98	0.65

Final actinide composition was calculated for every cycle and then the quantities of plutonium and depleted uranium to be added to the fuel were determined. The behavior of fast and thermal reactors was compared by following parameters: plutonium enrichment of the fuel; average value of neutron flux; share of the fissioning isotopes in plutonium; concentration of minor actinides. The results are presented in Figs. 1-6 as a function of total burnup for all cycles (which may be more than 100 per cent if the number of cycles is large enough - more than 15 for fast reactors and more than 25 for thermal reactors which refers to rather long periods of time).

These results demonstrate that:

- enrichment grows slower in fast reactor, almost stabilizing asymptotically; it's important because enrichment in thermal reactors is restricted by safety requirements at about 10 per cent;
 - average flux is practically stable in fast reactor but it's almost halved in thermal reactors and this decrease is to be compensated by enrichment;
 - degradation of Pu isotope composition is much faster in thermal reactors;
 - accumulation of neptunium is almost identical in both reactors up to 50 per cent burn up and then americium decay will increase it in TR (burn up is four times faster in fast reactors);
 - accumulation of americium and especially of curium is more intense in TR.
- These calculations were done for FR without breeding blankets.

2.2. The effects of plutonium composition and breeding ratio value on recycling of the actinides in fast sodium cooled reactors of BN type.

Multiple recycling of plutonium and MA in sodium cooled fast reactors BN-800 with highly Pu-enriched MOX-fuel was considered for the cases of civil Pu (BR=1.0 and 1.3) and weapon-grade Pu (BR=1.0) with irradiation cycle of 420 days. Following values were calculated:

- initial enrichment and final burn up in all three sub-cores for every cycle;
- plutonium isotopic composition at the beginning of every cycle;
- actinides concentrations at the end of every cycle.

Some of the results are illustrated by Figs.7-12. Following conclusions may be made:

1. There is prominent difference in the evolution of Pu isotopic composition during the recycling in converter and breeder modes: plutonium quality (the concentration of fissionable isotopes Pu-239 and Pu-241) is degrading significantly in converter mode while it is even improving slightly in the breeder mode, first of all due to accumulation of Pu-239 (see Fig.7) because very high grade plutonium is produced in the radial blanket (more than 96 per cent of Pu-239).
2. Minor actinides pile up slower in the breeder mode as may be seen in Figs.9-10 for the case of the most important nuclides Am-241 and Cm-244.
3. Switching to weapon-grade plutonium reduces accumulation of MA in converter mode drastically (see also Figs.9-10).
4. Tendencies mentioned in the points 2-3 are consequences of the reduced concentration of the key nuclide Pu-241 in the fuel (see Fig.8).
5. Practically all accumulation of Am-241 in plutonium fuel is due to the decay of Pu-241 in the load which may be seen in Fig.11 where the mass of Am-241 Vs Pu-241 mass in the load is shown. These masses are practically directly proportional both in for civil and weapon-grade plutonium.
6. Strong correlation exists also between the accumulated mass of most important curium isotope Cm-244 and Pu-242 concentration in the fuel load (Fig.12).

2.3. On combining thermal and fast reactors in Pu utilization and MA transmutation.

Various possibilities to create a combined system of fast and thermal reactors capable of recycling all the plutonium and MA are analytically explored now. Some of the latest results [23] are outlined below. The system considered includes two types of reactors: VVER-1000 with either uranium or 30 per cent MOX fuel and BN-800 with various cores. Basic parameters of the systems and some results of the calculations are given in Tables 7-9.

Table 7. Parameters of BN-800 cores.

Model number	Parameters of the model	Volume share of the fuel	Average Pu enrichment, per cent	Actinides burn up, kg/GWe year
1.	Uranium fuel with high Pu enrichment	0.29	37.0	57
2.	Inert matrix fuel without U-238	0.10	100	110

Table 8. Equilibrium fuel composition in VVER-BN system (kg/t).

Nuclide	Model 1				Model 2	
	Burnup 10%		Burnup 20%		Burnup 50%	
	Tc=1year	Tc=3 years	Tc=1 year	Tc=3years	Tc=1 year	Tc=3 year
U-235	2.4	2.3	2.1	2.1	-	-
U-238	564.1	575.9	521.4	516.4	-	-
Pu-238	12.3	14.7	13.5	15.0	46.6	53.5
Pu-239	151.4	153.8	170.1	171.6	249.0	250.1
Pu-240	137.6	140.3	153.6	155.5	358.7	358.3
Pu-241	30.0	26.6	38.1	35.6	80.2	67.9
Pu-242	38.6	38.4	47.3	47.4	118.6	119.0
Np-237	6.0	6.3	8.0	8.2	17.1	17.8
Am-241	12.4	17.9	13.8	17.6	47.2	62.1
Am-242	0.7	0.9	0.7	0.8	3.0	3.5
Am-243	14.3	14.3	18.0	18.2	46.6	43.7
Cm-242	0.2	0.02	0.2	0.02	0.6	0.06
Cm-244	7.8	6.8	10.4	9.1	26.3	20.1
Cm-245	2.1	1.8	2.7	2.5	5.3	4.0

Table 9. The number of VVER-1000 reactors with the actinide output utilized by one BN-800.

BN model	BN burnup		
	10%	20%	50%
Model 1	1.5 (1.0)*	1.7 (~ 1.05)	2.6 (2.6)
Model 2			

* Figures in brackets refer to 30 per cent MOX fueled VVER-1000.

3. The neutrons available for transmutation of fission products.

The price and value of the neutrons used for transmutation may be estimated by considering the income provided by nuclear electricity production. The approach [24] based on such considerations is used below in a simplified form. These results were presented in [25]

Modifications in energy producing system decreasing sales of electricity is the price of achieving environmental goals. To get one neutron as a result of interaction of high energy proton with specially designed target one needs not less than 20 MeV of beam energy. Taking thermal efficiency of electricity production equal to 30 per cent and accelerator beam efficiency 40 per cent we conclude that production of one "external" neutron takes the energy almost equal to that released in an act of fission. Thus the production of 4.2 kg of neutrons in the proton target means the loss of income equivalent to selling electricity generated by 1 t of fissioned heavy metal, i.e. approximately 1 GWt(e) year. If every neutron transmutes 1 nucleus of Tc-99 then that's the price of transmuting 420 kg of Tc-99.

Not only "external" neutrons may be used for transmutation but also reactor excess neutrons not used for energy production. Their alternative use may be breeding of Pu, so their price is not less than the value of lost Pu.

In the case of actinides transmutation in equilibrium mode when every nucleus of fresh fuel is sooner or later fissioned after a chain of nuclear transformations consuming neutrons the price of the neutrons is not easy to determine.

Hybrid transmutation concept when accelerator-driven blanket both produces energy and multiplies neutrons is most popular now. In this case both "external" and "multiplied" neutrons are used to transmute FP. But it does not a priori mean that one "external" neutron provides transmutation of more than one fission fragment.

Let's consider the neutronics of FP transmutation in accelerator-driven blanket. Notations used: ν - the number of fission neutrons, Σ_f - macroscopic fission cross section of the fuel; Σ_T - macroscopic total absorption cross section; Σ_{tr} - macroscopic cross section of FP placed in the blanket to be transmuted; $k = \nu \Sigma_f / (\Sigma_T + \Sigma_{tr})$ - blanket multiplication; $k_o = \nu \Sigma_f / \Sigma_T$ - initial multiplication of the blanket free from FP. The maximum number of fissions initiated by one external neutron in the blanket is $\phi = k / \nu(1 - k)$. The share of neutrons absorbed by FP being transmuted is

$$\eta = \Sigma_{tr} / (\Sigma_T + \Sigma_{tr}) = 1 - k/k_o \quad (1)$$

This definition differs from that of Takahashi [26] by a factor $(1 - k/\nu)$ because in [26] the share of absorptions used is $\eta_{FP} = \Sigma_{tr} / (\Sigma_T - \Sigma_f + \Sigma_{tr})$ which does not provide direct information on the share of neutrons used for transmutation.

The number of FP transmuted per one "external" neutron (the transmutation "value" of external neutron) is the product of η and the number of multiplied neutrons:

$$\tau = \eta(1 + \phi \nu) = (1 - k/k_o)/(1 - k) = 1 - \rho / \rho_o \quad (2)$$

where $\rho = (k - 1/k)$ is blanket reactivity. Let's now consider different reactivities of initial blanket (without FP added). If it were critical, $k_o = 1$, $\rho_o = 0$, we get $\tau = 1$, i.e. each "external" neutron provides transmutation of one FP just compensating added absorption. If initial blanket is subcritical ($k_o < 1$) then $\tau < 1$, i.e. "external" neutron transmutes less than one fragment in spite the multiplication in the blanket. And, finally, $k_o > 1$ results in $\tau > 1$.

If some quantity of FP is placed in initially supercritical blanket, making it precisely critical, the blanket will operate in reactor mode transmuted $1 - 1/k_o$ of fragments per every "internal" neutron, $\phi = k / \nu(1 - k)$ fissions will transmute $\tau^* = k(k_o - 1) / k_o(1 - k)$ fragments.

If the power of such a blanket, i.e. the rate of fissions, is equal to the power of subcritical accelerator-driven blanket, "combined" transmutation of FP in such a critical blanket plus direct transmutation by "external" neutrons will bring the same results as hybrid transmutation in subcritical accelerator-driven blanket with the same number of external neutrons: $\tau = \tau^* + 1$.

4. Criterion of radiation equivalence in environmental assessment of nuclear fuel cycles

Accumulation of highly radiotoxic nuclear wastes created a problem unique in the history of science - it's necessary to choose the direction of the development of nuclear technologies optimized both to ensure safety of our descendants in dozens and hundreds of thousands of years and to minimize radiological risks and economic losses for the present and nearest generations of nuclear electricity consumers who are not only to pay for those long-term safety measures but to learn how to handle and actually handle large volumes of radioactive materials taking risks and damages.

Now hazards of the nuclear power radwaste are estimated by comparison of its radiotoxicity with that of the excavated uranium ore (see, for example, [27,28]). This approach is typical for the use of traditional mineral resources - their volume reflects directly both their value and its environmental price. Nuclear power falls out of this picture in both aspects. Useful effect, i.e. the quantity of electricity produced, may be increased by almost two orders if we switch from today's open nuclear fuel cycle (ONFC) to closed one (CNFC).

We propose a modification of the assessment which is outlined below (see also [24]). In the estimates we shall assume the fission energy release equal to 200 MeV per act for all the actinides, differences in individual fission products (FP) yields for various actinides will be also neglected. In this approximation accumulation of 1 t of fission products corresponds to a little more than 1 GWt(e) year for standard thermal reactors. Consumption of uranium components and pile-up of the actinides are close for all major types of thermal reactors. Normalization of waste to 1 t of FP allows to compare hazards of various nuclear installations and cycles.

We shall use following mass notations: $M_{ore,i}$ - ore components, M_S - all spent fuel, $M_{F,i}$ - components of the fuel load, $M_{S,i}$ - components of the discharged spent fuel, M_{FP} - fission products, $\Delta_i = m_{S,i} - m_{F,i}$ where m_i are normalized to 1 t of FP. If specific "danger" of i-th nuclide is d_i then total danger of nuclides mixture is

$$D = \sum M_i d_i \quad (3)$$

What follows is true no matter what concept of "danger" is used, it may be mass, volume, activity, radiotoxicity, masses of diluting substances etc. Criterion of radiation equivalence of the spent fuel and excavated uranium usually used is the ratio of their dangers:

$$K_{S/R} = \sum M_{S,i} d_i / \sum M_{ore,i} d_i \quad (4)$$

Only about 0.7 per cent of natural uranium nuclei react in ONFC. More than 99 per cent of them were dislocated but their radiotoxicity was unchanged. Whether such "displaced" nuclei are taken into account or not almost does not influence the numerator value in Eq(4) but is very essential for the denominator - the mass of uranium excavated is by two orders larger than the mass of uranium transmuted, the ratio of their activities being about 40. If one applies the criterion (4) to ideal CNFC where all the uranium excavated is turned into FPs with some 200 times more electricity produced we get a paradox - most efficient use of natural uranium formally results in increased environmental danger because the accumulation of FPs per ton of natural U increases correspondingly. In practice lower specific consumption of natural uranium in CNFC is not taken into account in comparisons of the waste radiotoxicities of various NFCs. Such a "default" refusal to use the criterion (4) needs justification.

Let's now formulate a criterion of radiation equivalence which takes the specific ways of using raw materials in nuclear power into account. The idea is to compare the accumulated danger with the danger of transmuted (not only fissioned) uranium excluding "displaced" nuclides. The ratio of the dangers summed up over all the power and transmutation plants (index k) is

$$K_{W/U} = \sum_k M_{S,FP}^k \sum_i \Delta_{i,d_i}^k / \sum_k M_{S,FP}^k \sum_i \Delta_{ore,i}^k d_i \quad (5)$$

$\Delta_{ore,i}^k$ includes not only U isotopes but co-extracted products of their decay chains. Total energy produced is proportional to $\sum_k M_{S,FP}^k \sum_i \Delta_{FP,i}^k$. The numerator includes total accumulation of FPs and accumulation or burning of actinides, the denominator - total quantity of the transmuted uranium. In CNFC without FP transmutation only FP and irretrievable losses of actinides in reprocessing enter the numerator, and denominator includes 1 ton of U per every ton in the numerator. This criterion makes obvious the advantages of CNFC compared to ONFC in a very long run (thousands of years) when the main contribution to radiotoxicity is made by actinides, burnt in CNFC and accumulated in ONFC. This criterion is valid for the case of FP transmutation as well but it is not without deficiencies also - according to it burning of less active ^{238}U is 6 times more dangerous (due to small destroyed radioactivity) than burning of U-235.

The use of Eq (5) (accumulated danger/annihilated danger) instead of (4) (danger of spent fuel/danger of excavated uranium) increases the relative danger of ONFC correcting the traditional approach. It should be stressed that absolute indices of waste dangers do not depend on the choice of any of the two criteria.

Conclusions

Research and development efforts on transmutation should be aimed at the methods of closing nuclear fuel cycle efficient in a few senses:

- saving natural uranium by breeding secondary fuel;
- producing and directing to the radwaste less long-lived alpha-emitters;
- ensuring nuclear and radiological safety of all power producing plants and reprocessing installations;
- reducing proliferation dangers by keeping most of plutonium in spent fuel and strictly regulating quantity of separated plutonium.
- prompt introduction of fast reactors using Pu is vital for solving these problems;
- already nearest generation of new reactors with life-time expectancy of at least 50 years should be designed flexible to meet the requirements of steadily growing plutonium breeding when necessary;
- using of high-grade Pu in CNFC reduces the accumulation of minor actinides;
- useful energy produced should enter the denominator in any numerical criteria describing radiological hazards of NFC.

REFERENCES

1. GLOBAL'93. International Conference and Technology Exhibition. Future Nuclear Systems: Emerging Fuel Cycles & Waste Disposal Options. Proceedings. September 12-17, 1993, Seattle, Washington. Volumes 1-2.
2. GLOBAL 1995, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems. September 11-14, 1995, Versailles, France. Volumes 1-2.
3. Second International Conference on Accelerator-Driven Transmutation Technologies and Applications. 3-7 June 1966, Kalmar, Sweden. Abstracts.
4. E.V.Gai, B.K.Gordeev, A.L.Kochetkov, T.S.Milovidova, N.S.Rabotnov. The Closure of Nuclear Fuel Cycle: Actinides Balance, Safety and Fuel Provision Problems. The Eighth International Conference on Emerging Nuclear Energy Systems, ICENES'96, Obninsk, Russia, 24-28 June 1996. Book of Abstracts., p.4.
5. Suzuki T. Long-Term Logistic Analysis of FBR Introduction Strategy: Avoiding Both Uranium and Plutonium Shortage. In [2], v.2, p.1971.
6. V.I.Matveev. Choice and Preliminary Justification of BN-600 and BN-800 Reactors for Actinides burning and Its Fuel Cycle. PHYSOR-96 Paper M-290.
7. A.P.Ivanov, E.M.Efimenko, A.G.Tsykunov, G.G.Buiburin. Analytical Study on Optimization of Fast Reactors for U-233 Production and Plutonium and Minor Actinides Burning. PHYSOR-96 Paper SM-200.
8. A.P.Ivanov, E.M.Efimenko, A.G.Tsykunov. Fast Reactor Application for the Fission Products Burning. PHYSOR-96 Paper SM-150.
9. A.G.Kalashnikov, N.N.Novikova, Yu.G.Pashkin, V.V.Chekunov. Some Investigations of the Lead-Bismuth Cooled Fast Power Reactor for the Weapon-Grade Plutonium Utilization and the Actinide Burn-Up. The Eighth International Conference on Emerging Nuclear Energy Systems, ICENES'96, Obninsk, Russia, 24-28 June 1996. Book of Abstracts, p.132.
10. G.I.Toshinskiy. The Concept of Nuclear Power Development on the Basis of LMFBR Operating in the Open Nuclear Fuel Cycle. The Eighth International Conference on Emerging Nuclear Energy Systems, ICENES'96, Obninsk, Russia, 24-28 June 1996. Book of Abstracts, p.36.
11. T. Mukaiyama et. all, Actinide Integral Measurements on FCA for Evaluating and Improving Their Cross Section Data, 1985, NEACRP - A - 684.
12. S.P.Belov, J.C.Cabrillat et al.. Investigations of MA Transmutation Problem in Bench-Mark Experiments at BFS Facility with Neptunium in Fuel Composition. PHYSOR-96 Paper M-296.
13. L.P.Abagyan, N.O.Bazazyants, M.N.Nikolaev, A.M.Tsybulya. Group Constants for Reactor and Shielding Calculations. Energoizdat, Moscow, 1981(In Russian).
14. Yu.N.Shubin, V.P.Lunev, A.Yu.Konobeev, A.I.Dityuk. Cross Section Data Library MENDL-2 to study Activation and Transmutation of Materials Irradiated by Nucleons of Intermediate energies. INDC(CCP)-385, May 1995, IAEA, Vienna.
15. Yu.N.Shubin, V.P.Lunev, A.Yu.Konobeev. Semi-Empirical Systematics for (n,α) -reaction cross section. Nuclear Instruments and Methods in Physical Research. (1996), v.B108, pp.233-242.
16. A.Yu.Konobeev, V.P.Lunev, Yu.N.Shubin. Semiempirical systematics of (n,d) , (n,t) and (n,np) reactions cross sections at 14.5-15 MeV. IPPE Preprints 2491,2492,2494 (1996).
17. A.A.Veremeev, V.T.Gorshkov, B.F.Gromov, E.I.Efimov, M.P.Leonchuk. Preconceptual Design of Liquid Metal Targets with Diaphragm for Accelerator-Based Systems. [3], p.T/B7.
18. B.F.Gromov, Yu.I.Orlov, M.I.Sotov, V.A.Gulevsky, Yu.A.Musikhin. Studies of Windowless Liquid Metal Target with Lead-Bismuth and the Coolant Technology. [3], p.T/B25.
19. E.I.Efimov, D.V.Pankratov. Polonium and Volatile Radionuclides Output from Liquid Metal Target into Ion Guide and Gas System. [3],p.S/C3.
20. Yu.N.Shubin, A.V.Ignatyuk, A.Yu.Konobeev, V.P.Lunev, E.V.Kulikov. The Analysis of Energy Release, Beam Attenuation, Radiation Damage, Gas Production and Accumulation of Long-Lived Activity in Pb and Bi Targets. [3], p.M4.
21. E.I.Efimov, D.V.Pankratov, S.I.Ignatiev. Secondary Neutrons Yields by 0.8- and 1.6-GeV Proton Interaction with Pb and W thick Targets. [3], p.D47.
22. A.L.Kochetkov KARE code for the calculations of isotopes kinetics, radiational and environmental properties of nuclear fuel during its irradiation and cooling. IPPE preprint-2431, Obninsk, 1995.
23. V.M.Poplavskiy, V.I.Matveev, N.S.Rabotnov. Paper, presented to the Conference of Russian National Nuclear Society "New Power Technologies and the Role of the Nuclear Power, Fission and Fusion, September 14-18, 1996, Moscow.

24. H.Kuesters, H.W.Wiese, M.Salvatores. Transmutation of Actinides and Fission Products and Corresponding Fundamental Safety Aspects. In: Safety and environmental aspects of partitioning and transmutation of actinides and fission products. Proc. of a Technical Committee meeting held in Vienna, 29 November - 2 December 1993. IAEA-TECDOC-783. IAEA, January 1995., p.53.
25. E.V.Gai, A.L.Kochetkov, N.S.Rabotnov. Radiological Criteria, Potential and Limitations of ADTT at Closing Nuclear Fuel Cycle. [3], p.S35.
26. Takahashi, H., "Transmutation of High-Level Radioactive Waste by a Charged Particle Accelerator", in Proceedings of Global'93, International Conference on Future Nuclear Systems: Emerging Fuel Cycles and Waste Disposal Options, Seattle, WA, September 12-17, 1993, pp1293-1299.
27. Koch, L., "Minor Actinide Transmutation - a Waste Management Option", Journal of Less-common Metals, 122 (1986), pp371-382.
28. Adamov, E.O., Ganev I.C., Orlov, V.V., Attainment of Radiation Equivalence in Nuclear Power Radioactive Product Management, Nuclear Technology, v.104 (1993), N2, pp 233-241.

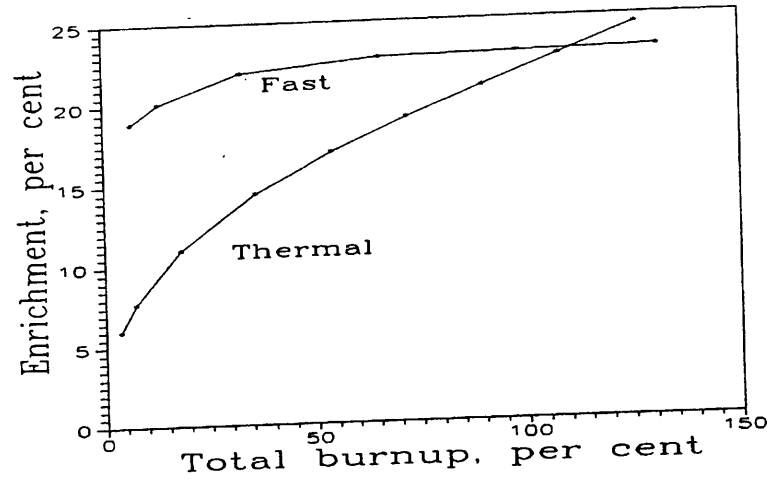


Fig.1. Increasing of enrichment in multiple recycling of actinides.

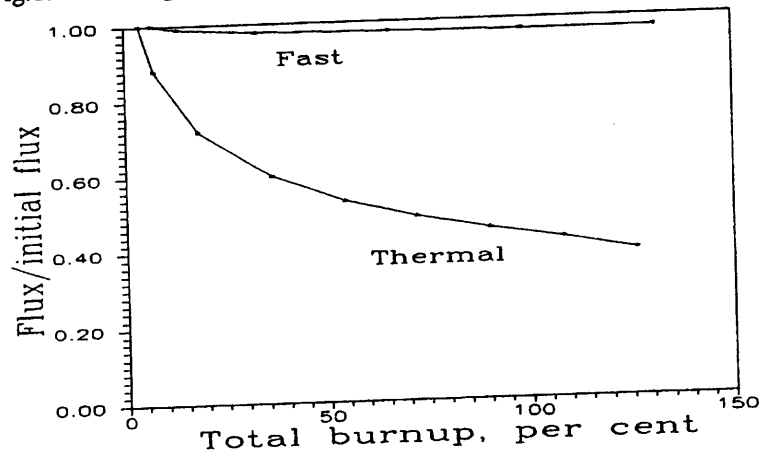


Fig.2. Flux degradation in multiple recycling of actinides.

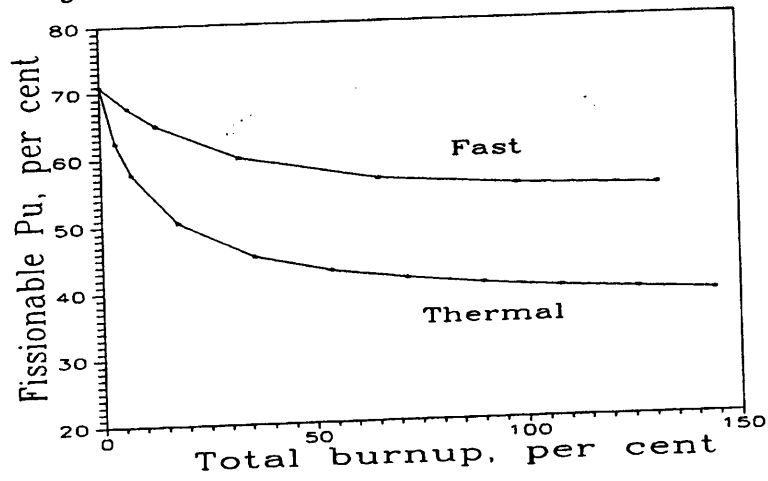


Fig.3. Degradation of Pu isotopic composition in multiple recycling.

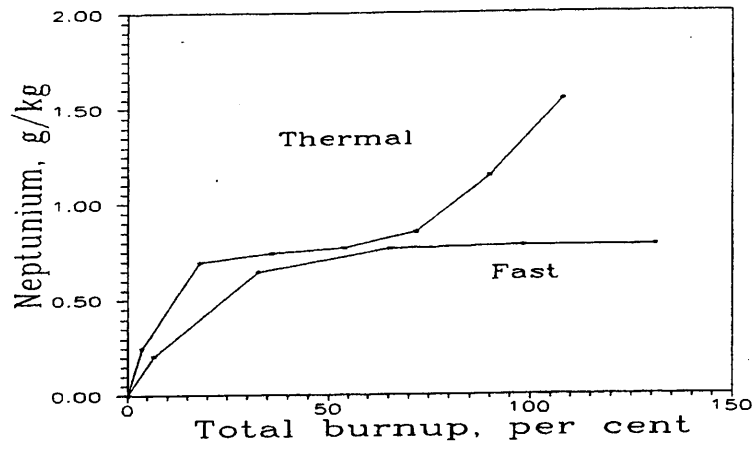


Fig.4. Accumulation of neptunium in actinides recycling.

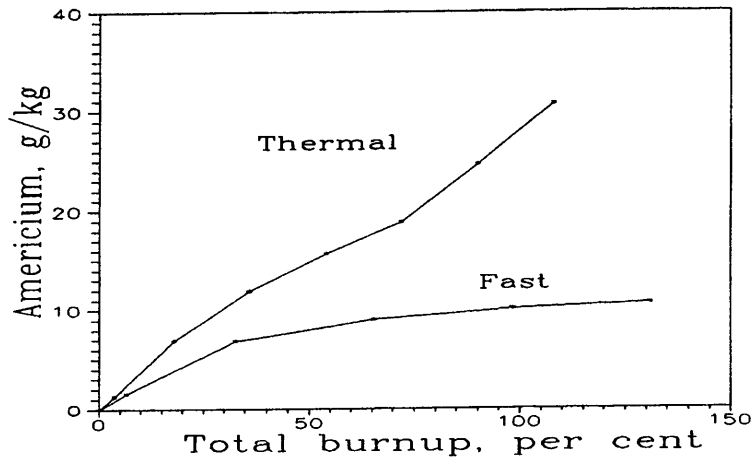


Fig.5. Accumulation of americium in actinides recycling.

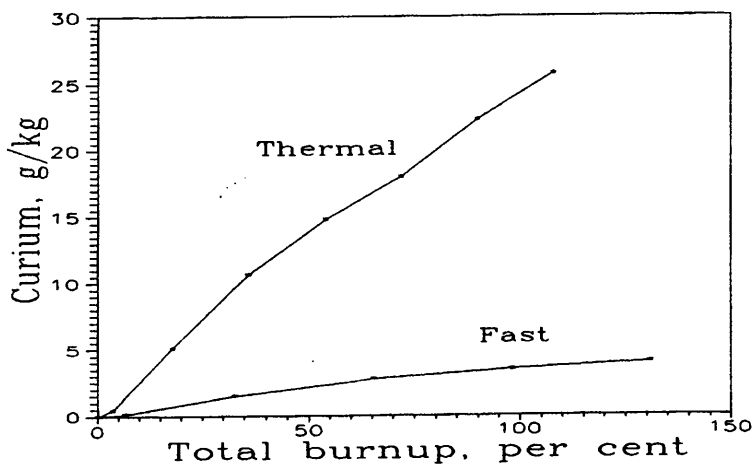


Fig.6. Accumulation of curium in actinides recycling.

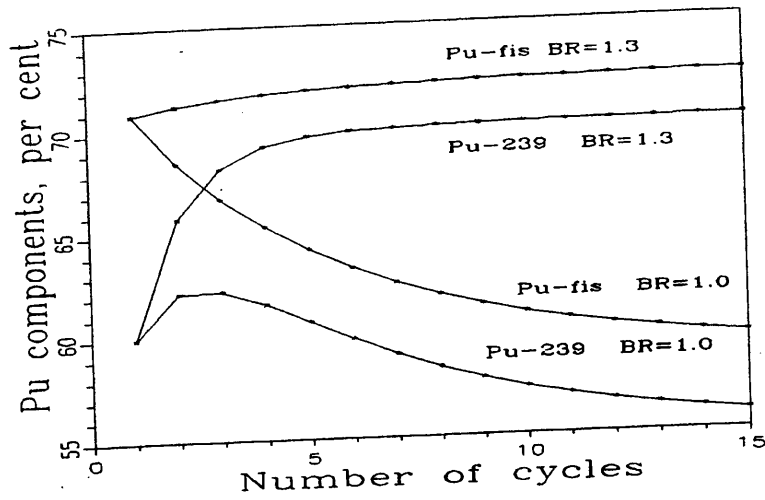


Fig.7. Evolution of Pu isotopic components in multiple recycling in BN-800 fast reactor at different values of breeding ratio.

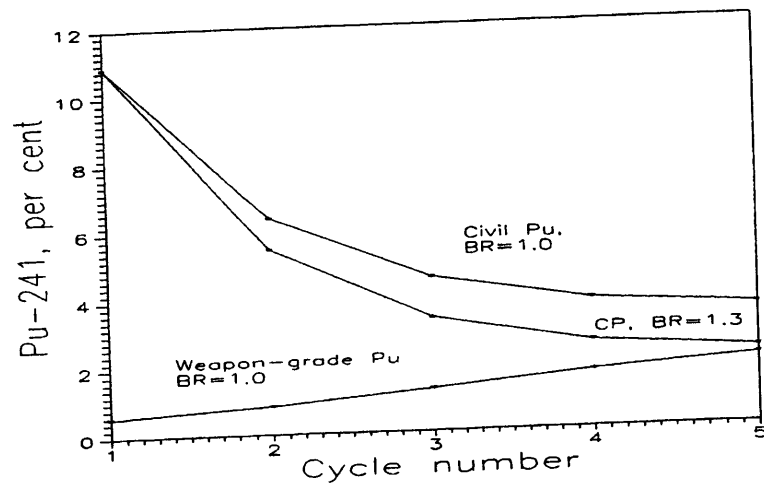


Fig.8. Total accumulation of Pu-241 in the core of BN-800 operating in various recycling modes.

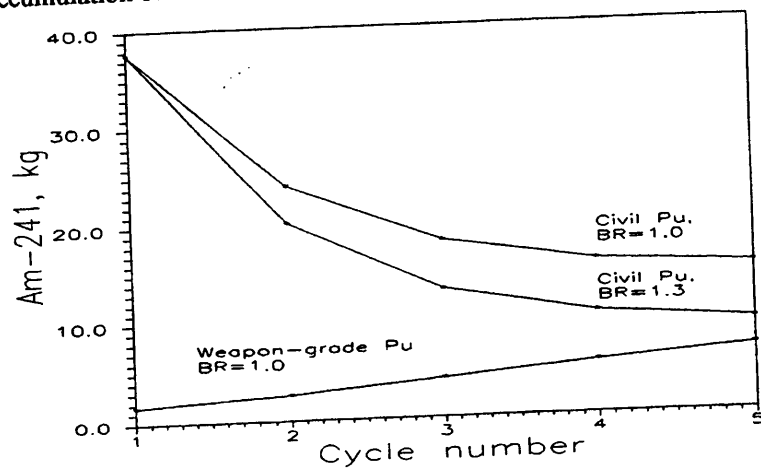


Fig.9. Total accumulation of Am-241 in the core of BN-800 operating in various recycling modes.

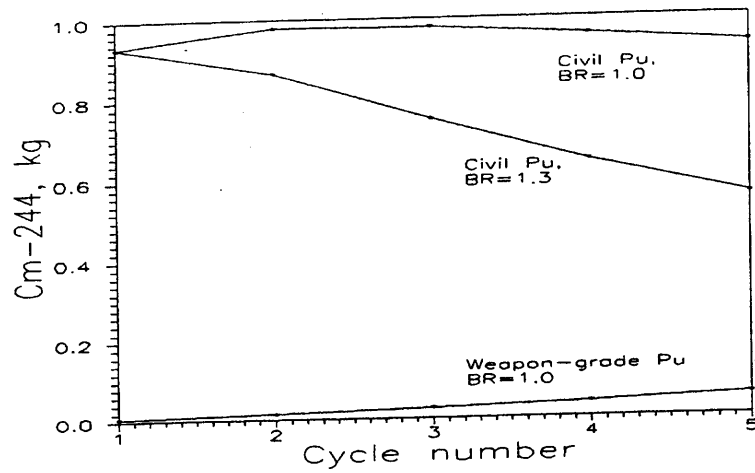


Fig.10. Total accumulation of Cm-244 in the core of BN-800 operating in various recycling modes.

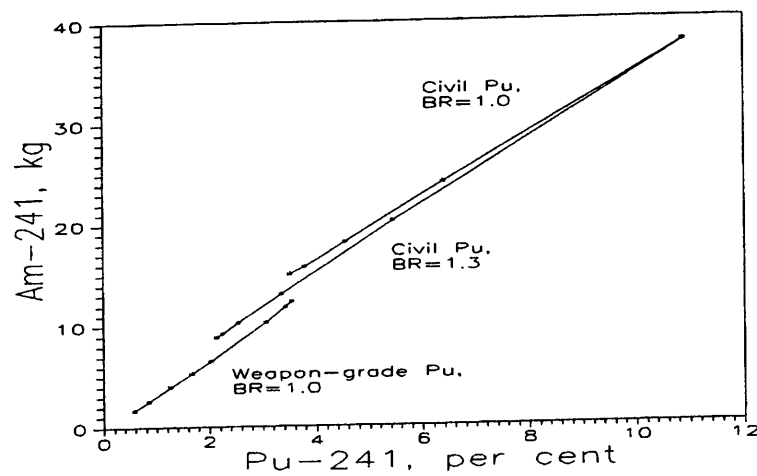


Fig.11 . Accumulation of Am-241 as a function of Pu-241 concentration in the fuel load of BN-800 during recycling of civil and weapon-grade plutonium in the converter mode (BR=1) and of civil Pu in breeder mode (BR=1.3).

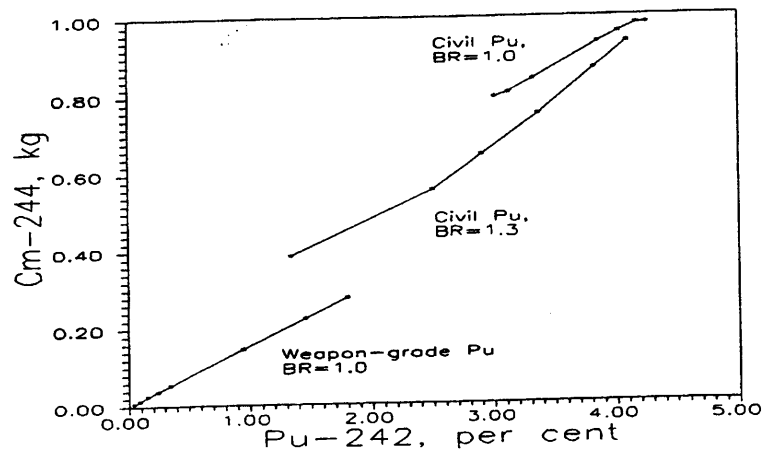


Fig.12. Accumulation of Cm-244 as a function of Pu-242 concentration in the fuel load of BN-800 during recycling of civil and weapon-grade plutonium in the converter mode (BR=1) and of civil Pu in breeder mode (BR=1.3).

ADVANCED FUEL RECYCLE SYSTEM CONCEPT TO REALIZE MINOR ACTINIDES RECYCLE

I.Nakajima, H.Ojima, S.Nomura
and T.Kawata

Power Reactor and Nuclear Fuel Development Corporation (PNC)

ABSTRACT

MA recycle has several benefits such as ease of waste problems and enhancement of proliferation resistance, but recovering MAs by means of aqueous process apparently needs more equipment, which requires more capital cost. The cost reduction by process optimization and simplification and integration of reprocessing and fabrication, which is one candidate goal of the Advanced Fuel Recycle System, is necessary to realize MA recycle.

In the present paper, PNC proposes an drastically improved recycle concept which is integrated system of reprocessing and fabrication. This concept mainly consists of a simplified PUREX process with single cycle extraction process flowsheet which gives rather low decontamination factors around 10^3 and simplified fabrication process such as vibro-packing. Such simplified extraction process can radically reduce number and volume of contactors, vessels, waste treatment components, and all auxiliary equipment which can brings forth the economical innovation.

The concept shows enough economical performance for application of MA recovery process.

1. INTRODUCTION

In compliance with the "Long-Term Program for Research, Development and Utilization of Nuclear Energy" issued in 1994 by the Atomic Energy Commission of Japan, PNC is expanding its development program for Advanced Fuel Recycle System as a new concept of fast breeder reactor (FBR) fuel cycle.

The primary objective of this program is to strengthen the performance of fuel cycle at various criteria. In the currently developed mixed oxide (MOX) of uranium (U) and plutonium (Pu) -fueled cycle, PNC recognizes that there are still issues to be substantially improved, particularly with respect to economics, generation of radioactive wastes both in terms of volume and quality, and international concerns about proliferation of nuclear material.

Recognizing the above-mentioned necessity of improvement, PNC believes that the following basic steps must be taken.

- (1) Improve the system's economics as a total fuel cycle system (system simplification)
- (2) Achieve Pu recovery without perfect purification
- (3) Make the most of the features of the fast neutron system. In addition to the breeding, pursue other function as MA (Minor Actinide such as Np, Am, and Cm) burning.

By following the above steps, the effective utilization of all actinides can be achieved, resulting in a reduction of the cumulative production of MAs, reduction of long-lasting toxicity in the high level liquid waste (HLW), and so on. This is the way to realize MA recycle.

Among several candidate system for Advanced Fuel Recycle System, PNC's main concern has been focused on the improvement of the system composed of the PUREX process and MOX fuel fabrication which have been well experienced by PNC through the past three decades.

2. FEATURE OF THE IMPROVEMENT

In the current commercial light water reactor (LWR) reprocessing, decontamination factor (DF) for fission products (FPs) in the order of 10^7 to 10^8 is required because of easy handling of the product U and Pu in the glove box for fuel fabrication.

On the other hand, we have to consider the trend of burn-up extension in the LWR system. The recovered Pu even with such high DF will have higher activity of gamma and neutron emission. This fact shows that the system of remote fuel fabrication should be essential in future.

If we allow the remote fabrication, the DF around 10^3 or even smaller are acceptable in the product materials for recycling it to the FBR, because FBR has the low neutronic sensitivity to its fuel material impurities. The FBR system does not require the fuel materials to be so pure like as the product from conventional reprocessing and such low DF can be attained easily by improved PUREX process with only one (single) cycle extraction.

In the conventional LWR reprocessing plant based on the PUREX technology using tri-butyl phosphate (TBP) as an extractant, the chemical separation process is composed of several extraction cycles in order to achieve high purity in both U and Pu products. Such multiple extraction cycles, usually consists of co-decontamination, partition of U and Pu and purification steps, become the multiple sources of aqueous and organic waste generation and then require quite a large capacity for the component to treat these waste. Figure 1 shows the ratio of liquid volume to be treated in each process of conventional reprocessing plant. It is apparent that around 70% or more are coming from partitioning of U and Pu and purification process.

Considering the single extraction process, we can eliminate all of partition and purification steps, which will result in the drastic reduction of total volume, i.e. 70% or more, and number of process component. Such reduction will lead to the proportional reduction in the required capacity of the waste processing and the reduction of waste itself. Thus the overall process is down sized and a significant reduction will be achieved on both plant capital and operating cost.

At present, the MOX fuel is usually fabricated in the form of sintered pellet at the facilities in the PNC Tokai Works as well as European countries. This fabrication method is featured with the powder preparation and pressing, sintering in furnace, chemical and physical inspections arraying pellets in a stack and inserting them into a cladding tube. Since most of these process depend on very precise mechanical handlings with complicated devices, they are not well fitted to the remote operation and alternative method are to be explored. Considering these facts, the gelation and vibro-packing method has been proposed as one of the methods suitable for remote fuel fabrication in Advanced Fuel Recycle System.

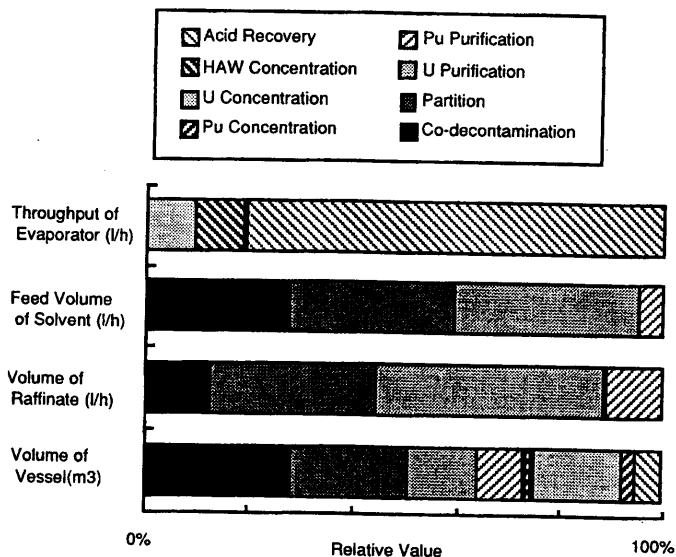


Fig.1 Volume comparison for each process in conventional reprocessing plant

3. DETAIL OF SINGLE CYCLE EXTRACTION PROCESS

One of the extremely simplified reprocessing flowsheet proposed as a part of Advanced Fuel Recycle System is shown in Figure 2, which is the improved PUREX process with single cycle extraction process as mentioned before.

In this case, both core and blanket fuel dissolved in nitric acid solution are treated by the process which consists of an extraction bank, a Pu and U co-stripping bank and a strip bank for residual U. The Pu and U co-stripping bank is made so that the Pu content is slightly higher than that required for core fuel fabrication. The process is also distinguished to extremely reduce amount of liquid waste by means of

- (1) eliminate scrubbing process, and
- (2) control process condition such as temperature, flow rate and so on in order to enable co-extraction of U and Pu without any reductant.

As for MA's behavior in this process, Np will be able to be recovered together with U and Pu if its valence in solution is adjusted in extractable form, but both Am and Cm will go to the raffinate from the extraction bank.

Further simplification will be made possibly by incorporating the supplemental technique to reduce heavy metal quantity to be fed to the extraction process in order to reduce aqueous and organic waste. One of the method under study is crystallization technique which is to collect, prior to solvent extraction, a large portion of uranium from the feed solution by crystallizing it as uranyl nitrate hexahydrate (UNH) at lower temperature. As the solubility of Pu is less than U and total amount of MAs and FPs are far below than the amount of U, only U crystallizes from the dissolved solution. After the crystallization, the residual solution with Pu/U ratio enriched to nearly 30% will be fed to the solvent extraction system which

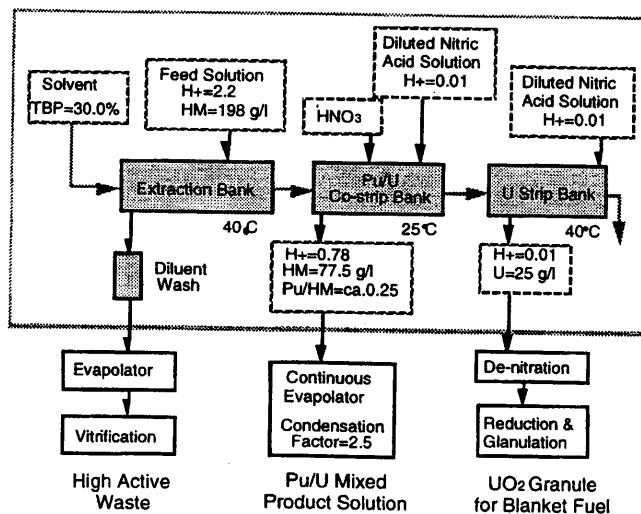


Fig. 2 Single-Cycle Extraction Flowsheet

consists of just an extraction and a stripping banks.

In either case, a major adjustment of Pu content in the product solution is to be made by the addition of recovered U before it is recycled for fuel fabrication.

It should be noted that the proposed process will completely eliminate the necessity for separation and handling pure Pu.

4. DETAIL OF REMOTE FUEL FABRICATION

Despite the automation of the process and inspection lines, radiation exposures to the workers tend to increase owing to occasional access for maintenance activities. In their aspect, a remote fuel fabrication process with an appropriate shielding is desirable or even necessary for the next generation MOX plants. And if we adopt the remote fabrication method for FBR fuel, we can drastically simplify the reprocessing process as discussed earlier.

Mixed solution of U and Pu with suitable concentration for MOX fabrication is fed to the gelation process to get particles with adequate diameter. Then particles are calcined and sintered to form granules. Finally granules are directly poured into cladding tube by applying vibration for dense filling.

In this case, most of the processes are fluidized, and therefore they are more suitable for remotization and easy to integrate into the reprocessing plant as its end process.

As shown in Figure 3, the system composed by gelation and vibro-packing can significantly shorten the fabrication process compared with the conventional process such as pelletization method. The inspection procedures will also be less complicated for the gelation process than in the pellet process.

5. EVALUATION ON ECONOMICSS

Since the major portion of electricity generation cost depends on the capital cost of the power reactor plant, significant effort is required for FBR plant to reduce its construction cost to become economically competitive. Also much effort to reduce the cost is needed for fuel cycle area which contributes about 20 to 30% of total electricity generation cost. About the fuel cycle cost comparison of LWR and FBR cycle, Figure 4 shows the relative effect of each constituents. As FBR fuel reprocessing has both functions of U purchase and conversion/enrichment in LWR cycle because of Pu recovery from spent fuel, it occupies larger portion in the fuel cycle cost than LWR fuel reprocessing. Innovation of reprocessing may brings forth larger cost effect. We believe the single cycle extraction process is one of the most effective method for this purpose.

In addition to the simplification and improvement of the process, the integration of reprocessing and fabrication processes into one fuel cycle plant will contribute to further reduction of the fuel cycle cost. It is often conceived that the remote handling might be more costly

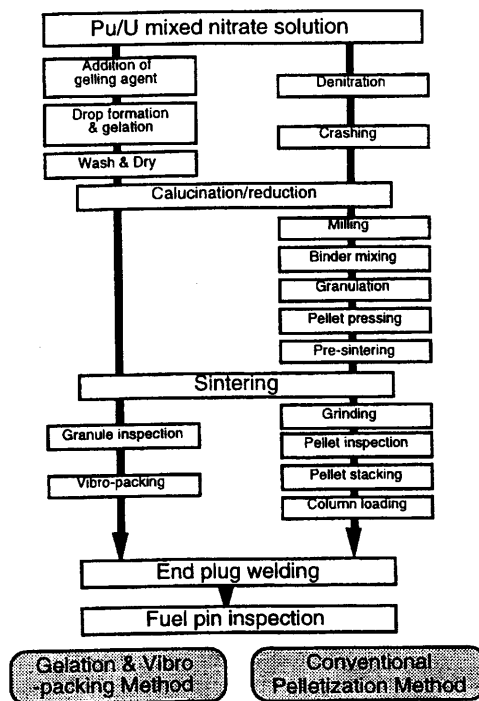


Fig. 3 Comparison of Fuel Pin Fabrication Process

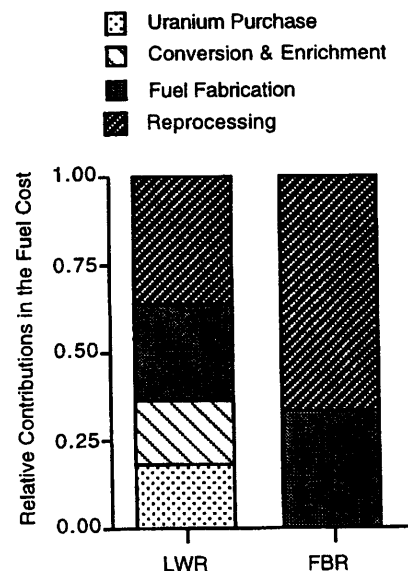


Fig. 4 Relative Comparison of Constituents in the Fuel Cost

than the automated directly handling. However, the remote fuel fabrication based on gelation and vibro-packing has a very good chance to achieve far better economy than the conventional pelletizing process, because the number of process steps in gelation is much smaller than in the pellet process preceded by the conversion process.

In Figure 5, a block flow diagram of an integrated fuel cycle plant based on Advance Fuel Recycle System is compared with that of conventionally separate reprocessing and fabrication plants. Based on a conceptual image of the Advanced Fuel Recycle Plant (AFRP), a preliminary cost evaluation has been carried out and compared with conventional reprocessing and fabrication plants with same throughput in Figure 6. As indicated in the Figure, the concept of AFRP will bring forth nearly 60% reduction in the plant capital cost for fuel recycling.

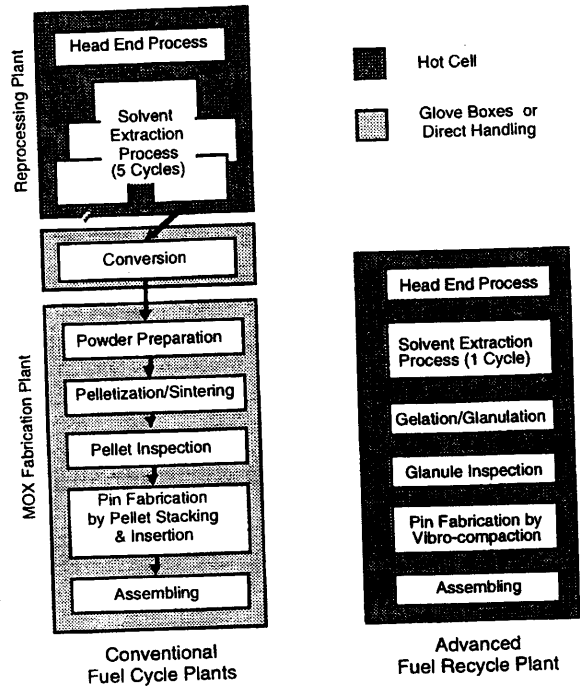


Fig. 5 Comparison of Fuel Cycle Plants

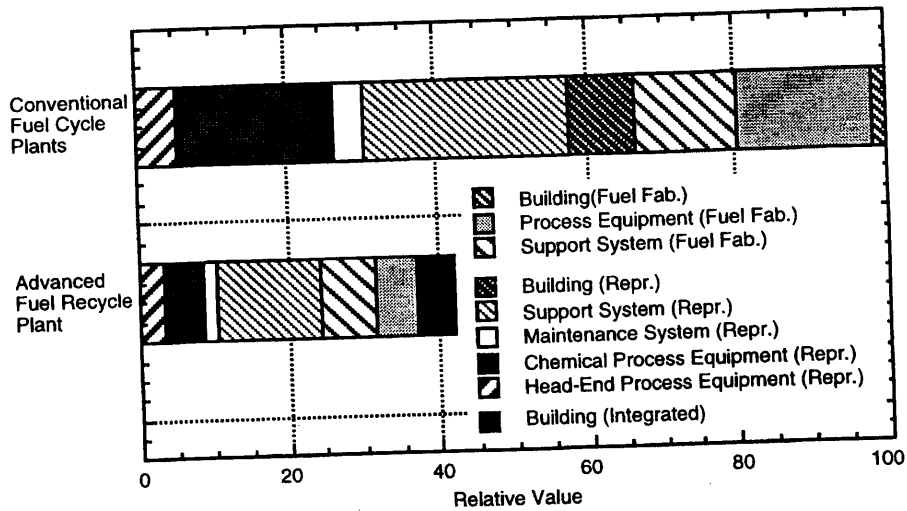


Fig.6 Capital Cost Comparison for Fuel Cycle Plants

6. CURRENT STATUS TOWARD MA RECYCLE

As the drastically improved recycle system signifies good performance on the economics, the addition of MA recovery system by means of aqueous process, that is TRUEX process in which CMPO is used as an extractant for MAs, may be acceptable, taking into account of merits of MA recycle such as easing the waste problem and strengthening the proliferation resistivity.

The cost gain due to addition of MA recycle function to the AFRP is estimated around only a few percent up. Of course, the MA recovery process such as TRUEX process should be reasonably improved.

It has been examined by TRUEX process that MA nuclides can be recovered almost 100% from the PUREX process raffinate generated from FBR spent fuel reprocessing test in the Chemical Processing Facility (CPF) in Tokai Works, PNC. Besides this, process condition to co-extract Np with Pu in PUREX process has been confirmed. Now, CPF is under modification to carry out more effectively the examination of the proposed single

cycle extraction process together with the basic study on the crystallization method focusing on the Pu solubility and the TRUEX process modification.

The possible recovered amount of MA from CPF is far below for conducting systematic irradiation test of MOX fuel containing MA nuclides, so that equipment to recover Am, which accumulates in the plutonium as a product from conventional LWR reprocessing plant, is going to be operation in near future at the Plutonium Fuel Development Facility (PFDF) in Tokai Works.

Enough amount of Am for irradiation test will be prepared into several pins at the cell of Alpha Gamma Facility (AGF), in Oarai Engineering Center, PNC, by remote technique to ensure workers exposure at low level. Irradiation test on MA contained fuel will be conducted at the Fast Experimental Reactor "Joyo" after getting the commission. It is expected that the test will be initiated around 2001.

SCENARIOS OF PLUTONIUM AND MINOR ACTINIDE MANAGEMENT AT EQUILIBRIUM

M. DELPECH, J.P. GROILLER, M. SALVATORES, J. TOMMASI, A. ZAETTA
CEA - Nuclear Reactor Directorate - Cadarache

A. HARISLUR, H. MOUNEY, M. ROME
EdF - Direction de l'Equipement

ABSTRACT

Any strategy related to radioactive waste management has to provide first a consistent answer on the Plutonium management. In fact, Plutonium build-up with standard PWRs should be accounted for and, if reprocessing is adopted, long-term solutions have to be found to use Plutonium in an effective way, and to minimise the associated minor actinide build-up.

The present paper gives the results obtained for different scenarios related to a nuclear power park of 60 GWe.

These results are given to show main trends and features, but do not account for detailed, economic and technological feasibility aspects, that will be the aim of future work.

1. INTRODUCTION

Any strategy related to radioactive waste management has to provide first a consistent answer on the Plutonium management. In fact, Plutonium build-up with standard PWRs should be accounted for and, if reprocessing is adopted, long-term solutions have to be found to use Plutonium in an effective way, and to minimise the associated minor actinide build-up.

In the frame of the studies required by the French Parliament law of 1991 on Waste Management, and according to the requirements of the National Commission (CNE) in charge of Evaluating these studies, the CEA Nuclear Reactor Directorate has launched a wide range of assessments on different scenarios of Pu utilisation in different reactor types (PWRs, FRs) and on the consequent approaches to the residual minor actinide management, and in particular to specific options to transmute them. The goal is to help feasibility assessments, to quantify fuel cycle characteristics and eventual benefits for deep storage.

This type of far-reaching studies is the closest to what can be considered a full system study, related to Plutonium management and partitioning/transmutation, and are an essential part of the SPIN program.

The present paper will give results and present trends.

2. THE SCENARIOS CONSIDERED IN THIS STUDY

The following scenarios have been characterised :

- Open cycle scenario.
- Pu recycling in light water reactors. Two major options for PWRs are considered : a) 100 % MOX-PWRs with a high moderator-to-fuel ratio, RMA, b) PWRs with MOX fuel on a ^{235}U enriched support, with standard moderator-to-fuel ratio.
- Pu recycling in PWRs (RMAs) followed by FR (CAPRA-type) utilisation for Pu multirecycling. Two hypothesis are considered : a) Only one recycle of Plutonium in PWRs, b) Two recycles of Pu in PWRs. Both cases attempt to consider delays in the introduction of FRs in a nuclear power park.
- In two cases : a) Full Pu recycling in PWRs, b) One recycle of Pu in a standard PWR followed by multirecycling in FR ; the explicit management of separated Np, Am, and Cm is considered.

All PWRs have a burn-up of 55 GWd/t, with fuel reloading frequency equal to six. Fast Reactors are of the CAPRA type (140 GWd/t burn-up, frequency 3).

For each of these seven scenarios, mass flow (Pu, minor actinides) throughout reactors and in the fuel cycle installations (reprocessing, fabrication) has been obtained.

All scenarios are relative to the same installed power (60 GWe, producing 400 TWhe/y).

Moreover, the detail of the reactor core characteristics is also given (enrichments at equilibrium, Pu vectors, detailed minor actinide inventories).

Finally the full Plutonium inventory at equilibrium is explicitly given. In terms of benefits for waste management in the storage, potential radiotoxicity and its evolution in time is presented for the scenarios where minor actinide transmutation is studied.

3. RESULTS OF THE SCENARIOS STUDIES

The results for the seven scenarios indicated above, are summarized in figures 1-7. Some comments on each scenario is given below.

3.1 Open cycle (figure 1)

This is the standard reference scenario. To be noted the high burn-up which has been chosen (and which is applied to all PWRs, whatever the fuel loading, in all subsequent scenarios).

The figure shows both the Uranium resources needed (in terms of annual mass flow) and both the resulting depleted Uranium (U-235 enrichment : 0.25 %) and the "wastes", which, in that case, include Plutonium.

3.2 Pu recycling in high moderation PWRs ($R_{mod} = 4$) (Figure 2)

In this scenario, all Plutonium is multirecycled in PWRs with a high moderator-to-fuel ratio, HMR, ($R_{mod} = 4$), which previous studies [1] have indicated as a particularly interesting option. Since the multirecycling is shown at equilibrium, one has an unlikely high Pu content in the fuel (~ 18 %). This scenario implies a 190 t MOX fabrication capability (to be compared to the present MELOX plant potential).

The HMRs represent ~ 20 % of the reactor park.

As far as resources, this scenario allows a ~ 20 % reduction of natural Uranium requirements.

As far as wastes, the Am and Cm production goes up of a factor of 4, with respect to the open cycle scenario.

3.3 Pu recycling in standard MOX PWRs with enriched Uranium support (figure 3)

In order to avoid potential problems related to the increase of Pu content in MOX PWRs (and the consequent potential problems related to reactivity coefficients), one can envisage a mixed enriched U-235-MOX fuel.

An equilibrium park is obtained with only one type of PWR. The fuel Pu content is ~ 2 %, and the U-235 enrichment is 3.8 % (to be compared to the standard 4.5 % value, see figure 1).

Multirecycling seems then feasible from a physics point of view. However two main drawbacks should be noted : a) Fuel fabrication costs will probably grow significantly, and MOX fuel fabrication plants should account for the annual need of 880 tons (\approx 7 MELOX plants) ; b) The Cm produced in this scenario goes up about a factor of 2.

In fact, the U-235 dominated core spectrum with a high thermalisation, favours the Cm production (the reduced Am production is an extra source of Cm build-up).

3.4 Scenarios with fast reactors for Pu recycling (figures 4 and 5)

Both scenarios of figures 4 and 5 account for the use of CAPRA-type fast reactors [2]. This seems to be still the most promising option, in view of the difficulties encountered in both the full-PWRs scenarios described above. For both scenarios, it has been considered realistic to envisage one or two recyclings of Plutonium in standard MOX-PWRs. The difference between the two scenarios is then represented by the delay in introducing fast reactors into the reactor park and the relative proportion of reactor types in the park (the fast reactor share is never higher than ~ 18 %). In fact, two recyclings of Pu in PWRs is envisaged in the scenario of fig. 5.

As far as resources, both scenarios allow a 30 % decrease of the natural Uranium supply requirement. In terms of MOX fabrication for both PWRs and FRs, one MELOX-type should be enough for the scenario of fig. 4. More fabrication capability is obviously required, if two recycling have to be foreseen in MOX-PWRs (fig. 5).

The waste production in these types of reactor parks, is close to that of the reactor park with HMRs (see figure 2). In fact, the potential benefits of using fast reactors, are reduced by the limited amount of these

reactors in the park and by the fact that the Pu recycled in fast reactor has already been degraded by one (or two) recycling(s) in PWRs.

Finally, it should be noted that the Pu content at equilibrium in the CAPRA-type reactors is close to 50 %, which is slightly higher of what is suggested in the CAPRA feasibility studies, but which can be easily adjusted to the required value (~ 45 %).

3.5 Scenarios with Minor Actinide (MA) recycling (figs. 6 and 7)

Minor actinides are recycled according to two scenarios which have been presented previously, namely a full PWRs scenario, with only one type of fuel loading (mixed enriched-U-235 and MOX fuel, shown in figure 3) and a MOX-PWR (one recycling) plus CAPRA-type reactors (shown in figure 4).

The mixed enriched-U/MOX PWRs have been chosen, since the MA recycling in PWRs generally worsens the core parameters [3], and, if an homogeneous MA recycling is chosen, one has to take that general trend into account. However, even in that case, the equilibrium composition (shown in fig. 6 : 4.1 % U-235, 2.6 % Plutonium, 1 % MA) can give rise to feasibility problems. As expected, the Cm production (see fig. 6) is very relevant with negative consequences on the physics parameters of the fuel cycle. Once more, it seems that the only reasonable way out to envisage a MA recycling in PWRs is the heterogeneous mode, and a specific scenario has to be envisaged, possibly with a temporary, long-term out-of-pile storage of Cm, or with a very long irradiation of MA targets, in order to fission them at a very high degree.

The scenario of figure 7 implies the use of FRs of the CAPRA-type. The relative fractions of the different reactor types are very close to those of fig. 4 (i.e. no MA recycling). The minor actinide recycling is of the homogeneous type for Np-237, and of the heterogeneous type for Am. Cm is separated and left to decay out-of-pile into Pu, and successively recycled [4].

This scenario does not present feasibility problems from the physics point of view. It has to be noticed that all the features of the specific, MA-burner CAPRA cores, are presently being experimentally studied in SUPERPHENIX [5] (high-Pu content fuel S/A in the CAPRA 1 and 2 irradiations, Np-bearing fuel in the homogeneous mode in the NACRE experiment. Am targets are foreseen to be introduced in SUPERPHENIX early 1998 : ECRIN experiment).

To optimise this scenario, the option of a once-through irradiation of Am targets is also being studied.

Finally, it is of interest to notice the overall amount of Am targets to be fabricated annually (\approx 5 tons).

4. THE POTENTIAL RADIOTOXICITY SOURCE IN THE STORAGE

It is easy, starting from the mass balances obtained in the studies described above, to evaluate the potential radiotoxicity source in a deep geological storage.

This parameter (when normalized to the energy which is produced), even if not to be taken as an absolute reference, is a simple way to characterize the activity inventory in a storage and its evolution with time. The Scenarios described in figs. 6 and 7 show a potential reduction of the radiotoxicity source term with respect to the open cycle scenario, of a factor which varies between 50 and 100, according to the time scale, when a decontamination factor of 0.1 % is applied to the Plutonium reprocessing and of 1 % to the MA partitioning (see fig. 8).

This factor is reduced to approximately 3 for all the scenarios, where only Pu is recycled, and MA are sent directly to the wastes (see fig. 8).

5. MASSES INVENTORY

All the previous discussion has been based on annual mass flows. However, an interesting point is to consider the total isotope masses which are present in the fuel cycle. These quantities will help to understand the implications of the equilibrium scenarios, in the case, for example, of the need to reduce drastically a nuclear power park.

Fuel cycle inventory is defined as the total isotope masses present in Reactors and in the different fuel cycle plants (reprocessing, storage, fabrication). We consider five years for cooling time before reprocessing and two years for fabrication operations.

For the different scenarios, masses inventories are given in the following table :

	Pu Inventory (tons)	Minor Actinides Inventory (tons)
Pu recycling in HMR (fig. 2)	430	/
Pu recycling in PWR with U235 support (fig. 3)	230	/
Pu recycling in CAPRA (once in PWR (fig. 4))	370	/
Pu recycling in CAPRA (twice in PWR (fig. 5))	380	/
Pu + M.A recycling in PWR with U235 support (fig. 6)	300	95
Pu + M.A recycling in CAPRA Pu once in PWR (fig. 7)	440	90

Scenarios with Pu recycling in dedicated Reactors (PWR or FR) show a quite similar amount of Pu inventory (around 400 tons).

For the scenario with Pu recycling in standard PWR with U235 support, Pu inventory is rather smaller about 200 tons. This is mainly due to the fact that in these case, the fissile isotope inventory necessary to reach Reactor's criticality is partially provided by the U235 support.

When Minor Actinides are recycled, the Pu inventory is increased by about 20 % to 30 % and Minor actinides inventory is slightly smaller than 100 tons for both scenarios.

REFERENCES

1. M. Salvatores et al. "Nuclear Waste Transmutation : physics issues and potential in Neutron fields, ENS Conf. Tel-Aviv, January 1994.
2. J. Rouault et al. "The CAPRA feasibility study", Proc. 3rd Int. CAPRA Seminar, Lancaster, 1995.
3. M. Delpech et al. "PWRs potential for Pu and Minor Actinide burning", ENC'94, Lyon, Oct. 1994.
4. J. Tommasi et al, Nuclear Technology, 111, 133 (1995).
5. P. Anzieu, R. Del Beccaro, "Pu burning and Actinide Transmutation in SUPERPHENIX", GLOBAL'95, Versailles, Sept. 1995.

FIGURE 1
PARK OF PWRs (60 GWe) - 400 TWhe/year
Burn-up = 55 GWD/t Frequency = 6

1. OPEN CYCLE

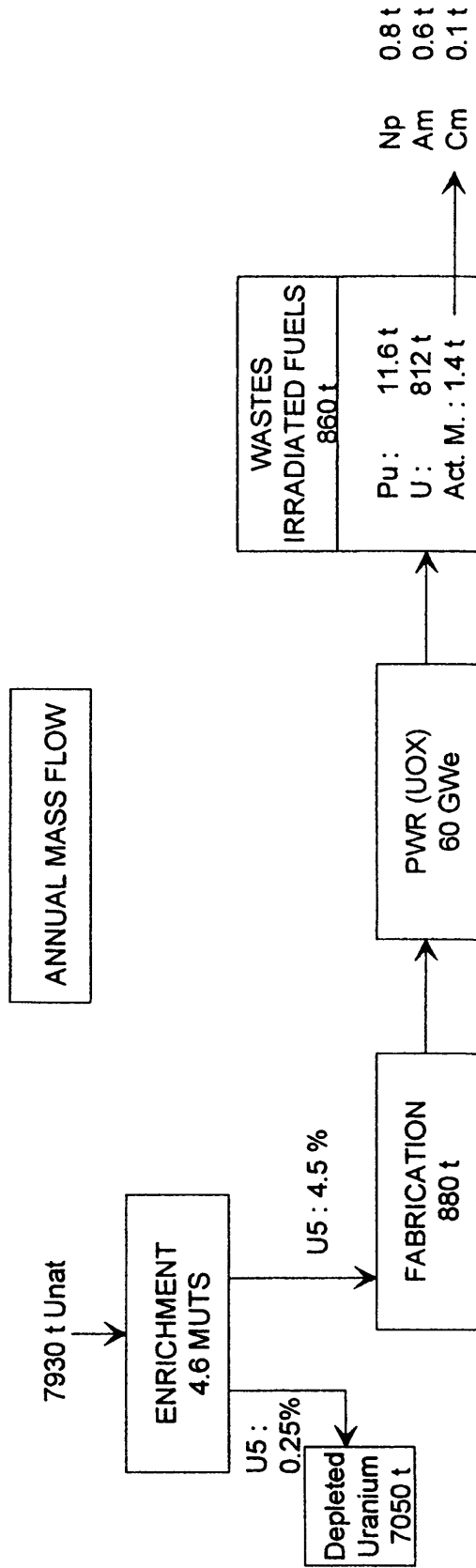


FIGURE 2
PARK OF PWRs (60 GWe - 400 TWhe/year)
EQUILIBRIUM STATE
BURN-UP : 55 GWD/t Frequency = 6

2. RECYCLE OF PU IN HIGH MODERATED REACTOR (Rmod = 4)

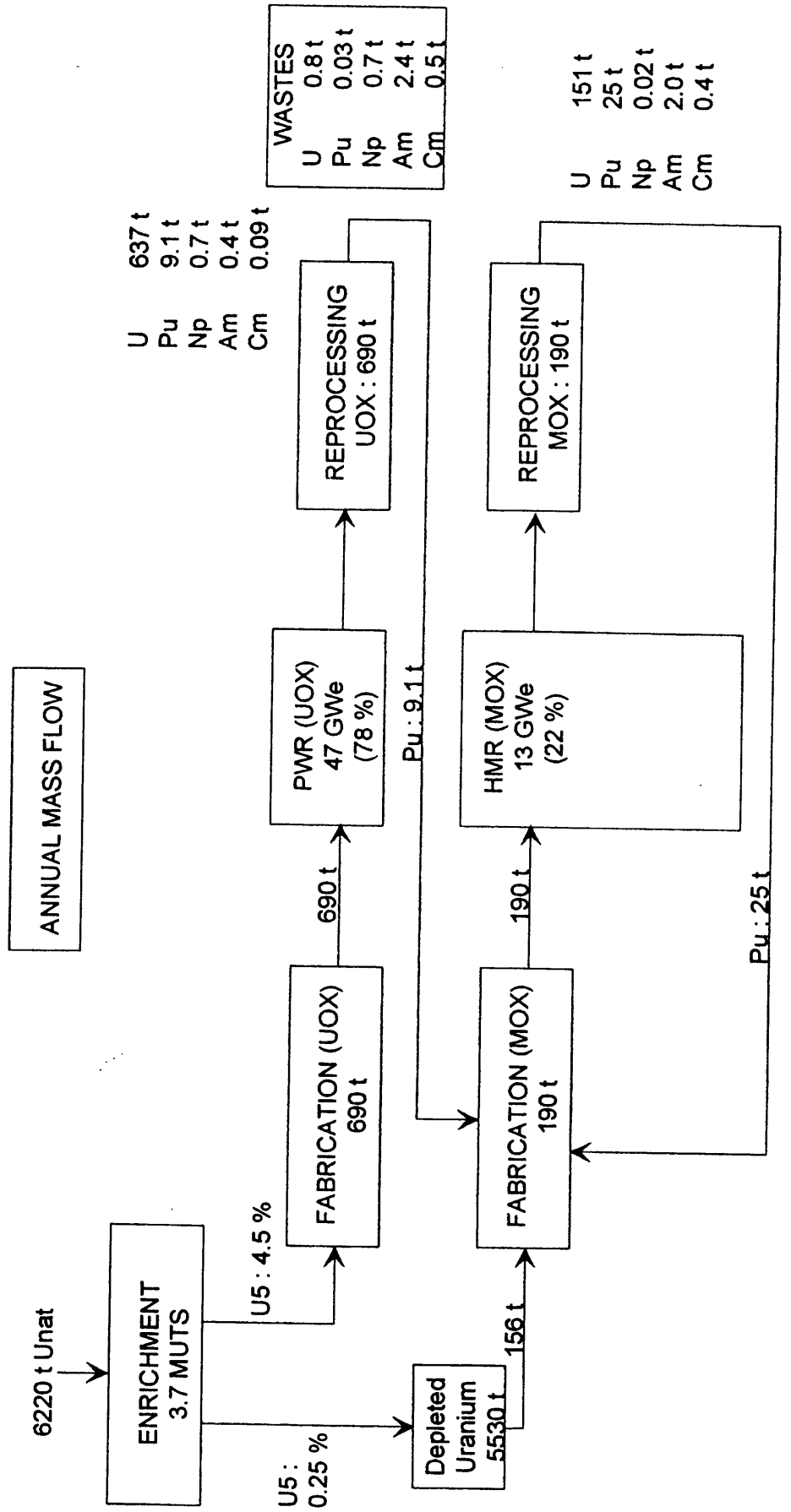


FIGURE 3
PARK OF PWRs (60 GWe - 400 TWh/year)
EQUILIBRIUM STATE
BURN-UP : 55 GWD/t Frequency = 6

3. RECYCLE OF Pu IN STANDARD MOX WITH ENRICHED URANIUM SUPPORT

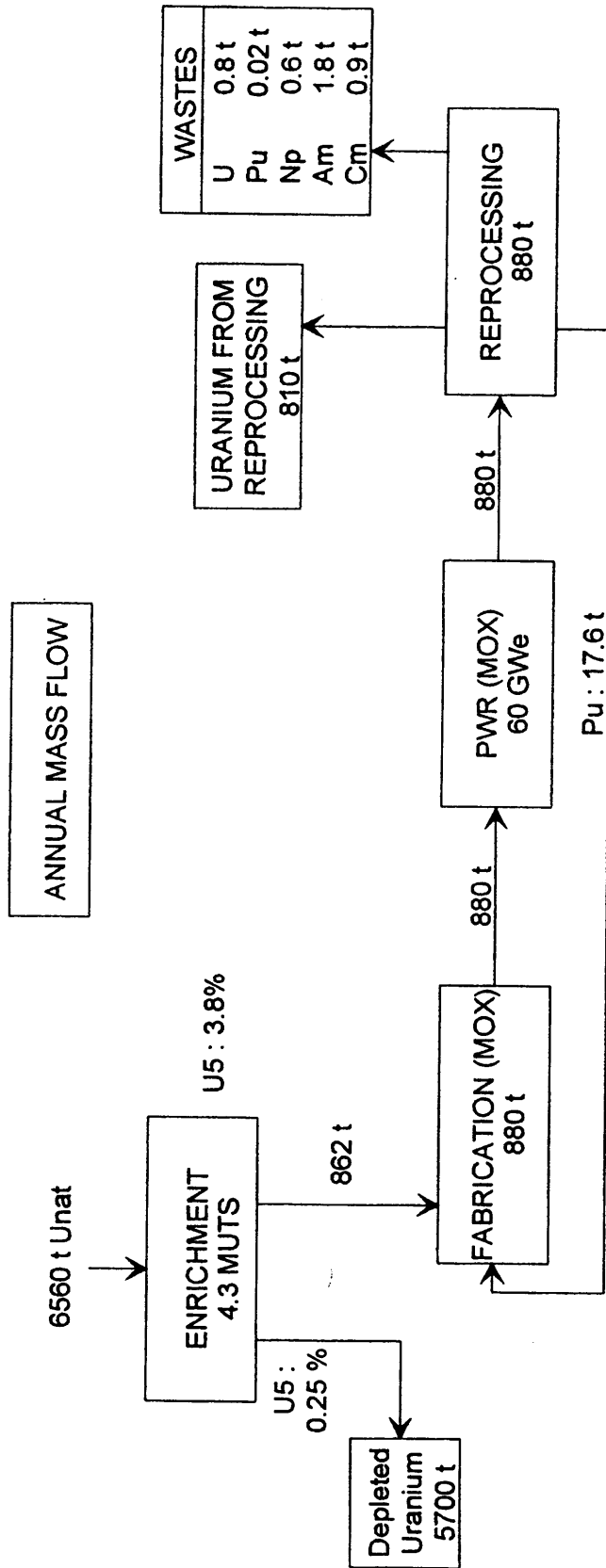


FIGURE 4
PARK PWR + CAPRA (60 GWe - 400 TWhe/year)
EQUILIBRIUM STATE
Frequency 6
Frequency 3
BURN-UP PWR = 55 GWD/t
BURN-UP CAPRA = 140 GWD/t
4. RECYCLE OF Pu : once in PWR and after in CAPRA

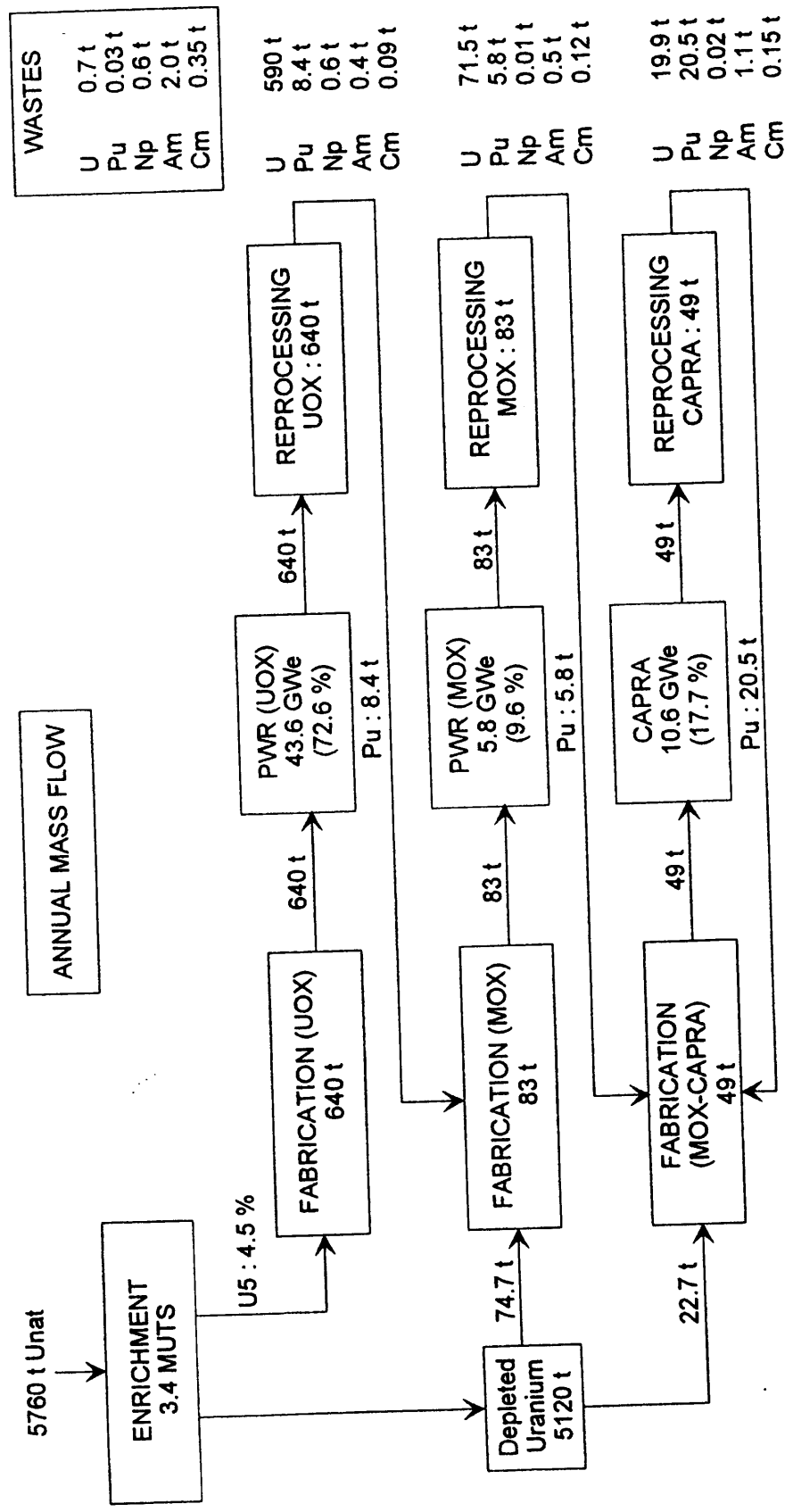


FIGURE 5
PARK PWR + CAPRA (60 GWe - 400 TWhe/year)
EQUILIBRIUM STATE
BURN-UP PWR = 55 GWD/t **Frequency 6**
BURN-UP CAPRA = 140 GWD/t **Frequency 3**
5. RECYCLE OF Pu : twice in PWR and after in CAPRA

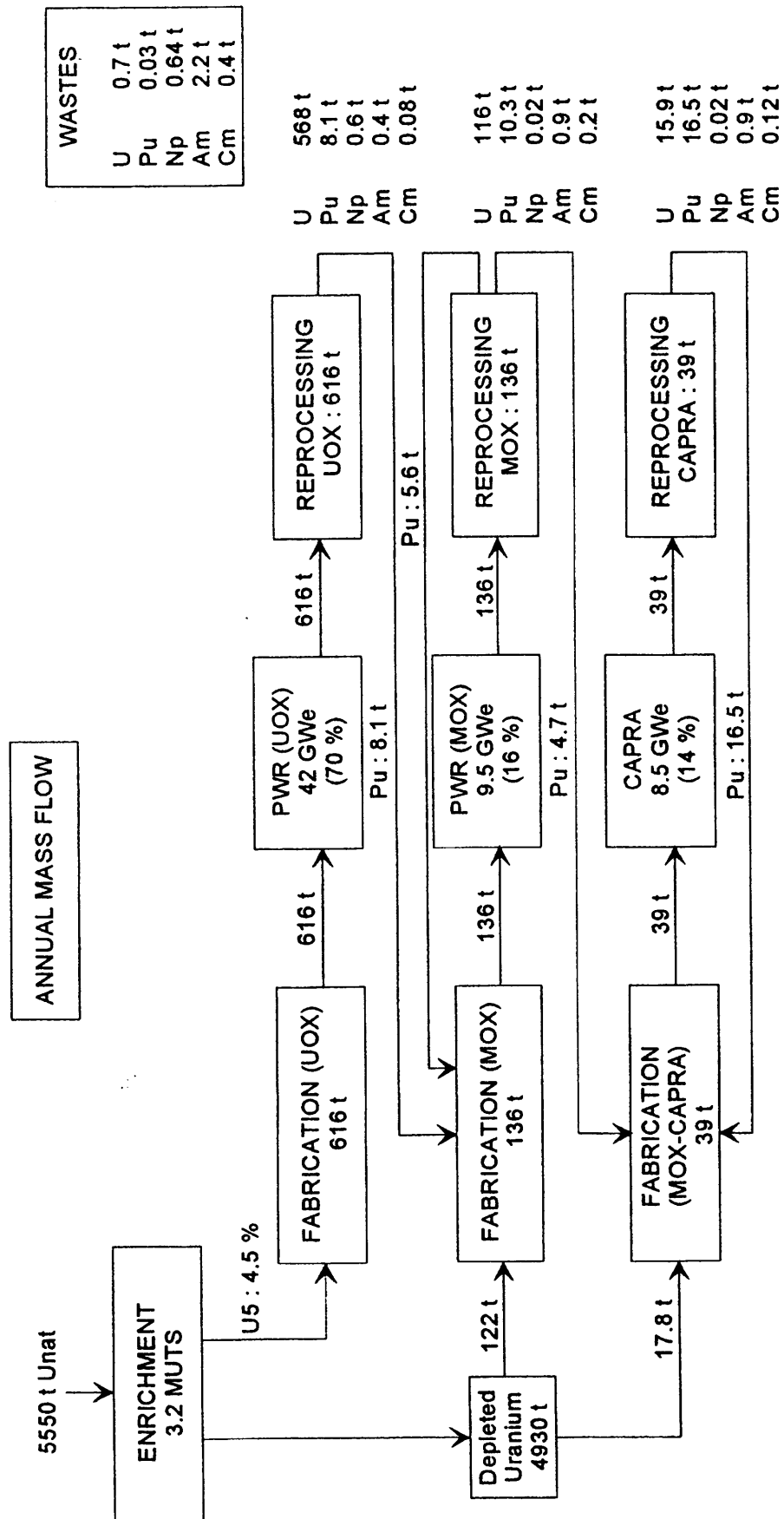


FIGURE 6
PARK OF PWRs (60 GWe - 400 TWh/year)
(55 GWD/t, Frequency = 6)
EQUILIBRIUM STATE

6. RECYCLE OF Pu + Np + Am + Cm in MOX PWR with enriched uranium support :

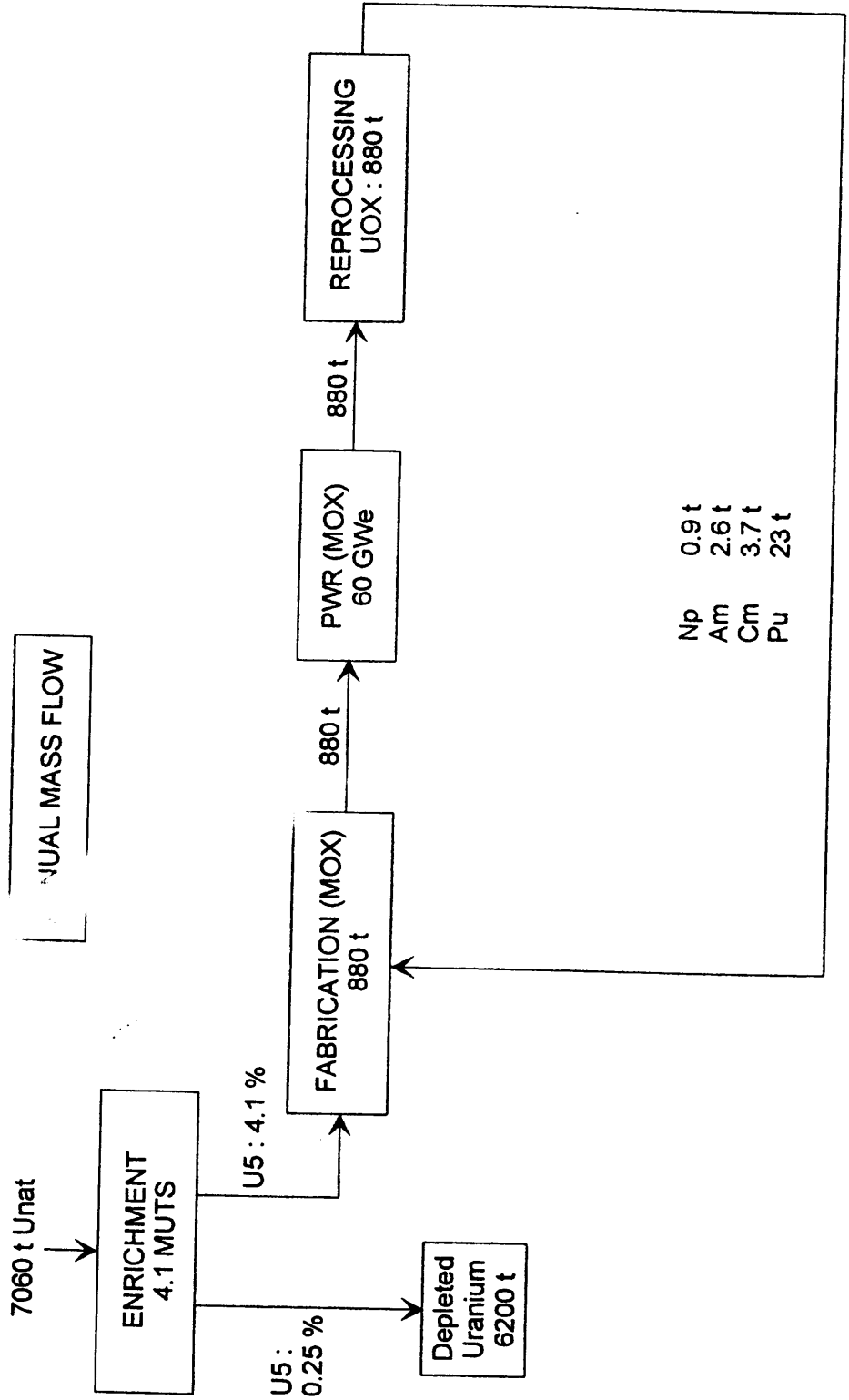


FIGURE 7
PARK PWR + CAPRA (60 GWe - 400 TWhe/year)
EQUILIBRIUM STATE
PWR = 55 GWD/t Frequency 6
CAPRA = 140 GWD/t Frequency 3
7. RECYCLE OF : once in Pu in PWR and after in CAPRA, Np + Am + Cm
in CAPRA : 5680 t Unat

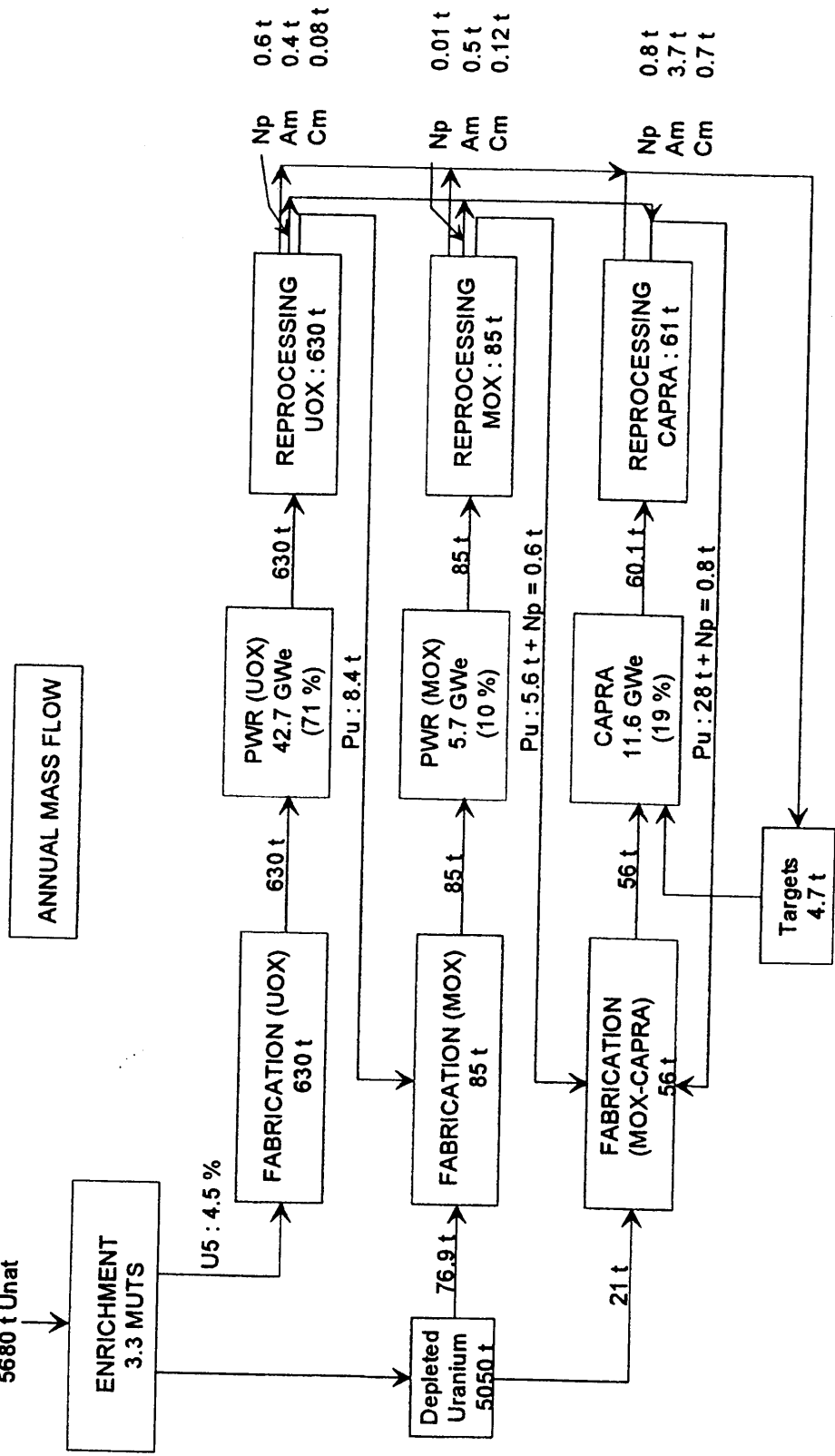


FIGURE 8

